

Universal Property of Quantum Measurements of Equilibrium Fluctuations and Violation of Fluctuation-Dissipation Theorem

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For macroscopic quantum systems, we study what are measured when equilibrium fluctuations of macrovariables are measured in an ideal way that mimics classical ideal measurements as closely as possible. We find that the symmetrized time correlation (symTC) is always obtained for such measurements. As an important consequence, we show that the fluctuation-dissipation theorem (FDT) is partially violated as a relation between observed quantities in macroscopic quantum systems even if measurements are made in such an ideal way.

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When temporal fluctuations of observables are measured in quantum systems, disturbances by the measurement play crucial roles [1–6]. Suppose that one measures fluctuation of \hat{A} by measuring \hat{A} at time $t = 0$ and subsequently at time $t > 0$. Then the disturbance by the first measurement affects the outcome of the second one. Consequently, fluctuation of \hat{A} , expressed by its time correlation, depends strongly on the way of measuring it. For example, the normal-order time correlation and antinormal-order time correlation are obtained, respectively, when a photon field is measured with a photodiode and a quantum counter [1–3]. This means, for example, that the former cannot measure the zero-point fluctuation [3].

Such strong dependence on measuring apparatuses should also be present in measurements of equilibrium fluctuations in macroscopic quantum systems. This may affect fundamental relations such as the FDT, which is a universal relation between response functions (that characterize responses to driving forces) to equilibrium fluctuations (that are expressed by time correlations) [7–12]. However, the disturbances by measurements were completely disregarded when deriving the FDT [8–12]. It is obvious that violent measurements of time correlations will lead to apparent violation of the FDT because of strong disturbances. Hence, one would expect that the FDT would hold only when certain ‘ideal’ measurements are made for time correlations. For classical systems, ideal measurements are those which do not disturb the system at all. For quantum systems, however, such measurements are generally impossible. Hence, ‘ideal’ measurements are those which mimic classical ideal measurements as closely as possible. It is expected that the FDT should hold when such ‘ideal’ measurements are made. Is this expectation true?

A similar problem exists in Onsager’s regression hypothesis [13, 14]. While Refs. [15, 16] claimed its consistency with the Kubo formula [10], other works claimed inconsistency [17–19]. To settle this controversy in an operational manner, disturbances by measurements should be considered, which were disregarded in these works.

In this paper, we study, using modern theory of quantum measurement [1–6], what is observed when ‘ideal’ measurements are made on equilibrium fluctuations. We find the universal answer that the symTC is always observed. As a striking consequence, the FDT and the regression hypothesis are violated as relations between observed quantities. Furthermore, we show that in the post-measurement states, unlike in the Gibbs states, expectation values of macroscopic observables evolve with time. Such states should be realized in experiments on equilibrium fluctuations.

Assumptions on equilibrium state.— We consider a d -dimensional macroscopic quantum system ($d \geq 1$), whose size is characterized by N (such as the number of spins). The pre-measurement state, represented by the canonical Gibbs state $\hat{\rho}_{\text{eq}}$, is its equilibrium state at finite temperature T ($= 1/\beta$).

We assume that the correlation between any local observables at two points \mathbf{r} and \mathbf{r}' decays faster than $1/|\mathbf{r} - \mathbf{r}'|^{d+\epsilon}$ ($\epsilon > 0$) with increasing $|\mathbf{r} - \mathbf{r}'|$. This assumption is believed to hold generally except in special regions, such as critical points, in the thermodynamic configuration space. Under this assumption, the static equilibrium fluctuation, $\delta A_{\text{eq}} = (\langle (\Delta \hat{A})^2 \rangle_{\text{eq}})^{1/2}$, of any additive observable \hat{A} is $O(\sqrt{N})$ [20–22]. Hereafter $\langle \cdot \rangle_{\text{eq}} \equiv \text{Tr}[\hat{\rho}_{\text{eq}} \cdot]$ and Δ denotes the shift from the equilibrium value, i.e., $\Delta \hat{A} \equiv \hat{A} - \langle \hat{A} \rangle_{\text{eq}}$. Furthermore, with some reasonable additional assumptions (see Ref. [23]), the ‘quantum central limit theorem’ [23–27] holds, which enables us to derive the following universal results. Its statement, discussion on related theorems [28–30], and details of the calculations are described in Ref. [23].

Quasiclassical measurement.— As discussed above, certain ‘ideal’ measurements, which mimic classical ones, should be made to get time correlations correctly. We call such measurements *quasiclassical*. To be definite, we define them as minimally-disturbing, homogeneous, and unbiased quantum measurements with moderate magnitudes of errors, as follows.

To observe fluctuation of an additive observable \hat{A} , its measurement error δA_{err} should be smaller than δA_{eq} .

On the other hand, δA_{err} should not be too small because otherwise disturbances by the measurement would be too large so that the measurement would be much different from classical ideal ones. Therefore, we consider measurements in which $\delta A_{\text{err}} = \varepsilon \delta A_{\text{eq}}$, where ε is independent of N . Although we are interested in the case where $\varepsilon < 1$, the following results hold also for larger ε (which occurs, e.g., when interaction with the measuring apparatus is weak [4]). Considering $\delta A_{\text{eq}} = O(\sqrt{N})$ as mentioned above, we require more concretely (i) the measurement operator (that gives the state just after measurement [5, 6]) scales as \sqrt{N} with increasing N . A quasiclassical measurement should also possess the following reasonable properties: (ii) Minimally disturbing, i.e., the only disturbance of the system is the necessary backaction determined by the probability operator [5]. (iii) Homogeneous, i.e., the measurement operator is a function of $\hat{A} - A_{\bullet}$, where A_{\bullet} is the outcome of the measurement. This implies, e.g., a reasonable condition that δA_{err} is independent of A_{\bullet} . (iv) Unbiased, i.e., $\overline{A_{\bullet}} = \langle \hat{A} \rangle_{\text{eq}}$, where $\overline{\dots}$ denotes average over many runs of experiments. From (i)-(iv), the measurement operator should take the form $f(\hat{a} - a_{\bullet})$ in terms of scaled quantities $\hat{a} = \hat{A}/\sqrt{N}$ and $a_{\bullet} = A_{\bullet}/\sqrt{N}$. Here, $f(x)$ is a real function independent of N such that $\int |f(x)|^2 dx = 1$, $\int x |f(x)|^2 dx = 0$. Hence, the probability density for getting a_{\bullet} is

$$p(a_{\bullet}) = \langle \{f(\hat{a} - a_{\bullet})\}^2 \rangle_{\text{eq}}, \quad (1)$$

and the post-measurement state for the outcome a_{\bullet} is [31]

$$\hat{\rho}(a_{\bullet}) = f(\hat{a} - a_{\bullet}) \hat{\rho}_{\text{eq}} f(\hat{a} - a_{\bullet}) / p(a_{\bullet}). \quad (2)$$

It seems reasonable to require also (v) $f(x)$ behaves well enough, e.g., it vanishes quickly with increasing $|x|$ (such as the Schwartz functions [32–35]; see Ref. [23] for details). Although (ii) requires also that f should be nonnegative up to an irrelevant phase factor [5], all the following results hold without this additional condition. Then, the measurement error δa_{err} ($= \delta A_{\text{err}}/\sqrt{N}$) is given by $(\delta a_{\text{err}})^2 = \int x^2 |f(x)|^2 dx = O(1)$, in consistency with $\delta A_{\text{err}} = \varepsilon \delta A_{\text{eq}}$. A typical f is gaussian [37], $f(x) = (2\pi w^2)^{-1/4} \exp(-x^2/4w^2)$ with $w = O(1) > 0$, for which $\delta a_{\text{err}} = w$.

Which time correlation is measured?— Although all the following results hold in the thermodynamic limit, we do not write $\lim_{N \rightarrow \infty}$ explicitly. Suppose that the macroscopic system was in a Gibbs state $\hat{\rho}_{\text{eq}}$ for $t < 0$, and that an additive observable \hat{A} is measured at $t = 0$. The outcome a_{\bullet} ($= A_{\bullet}/\sqrt{N}$) distributes according to Eq. (1), for which we have

$$p(a_{\bullet}) = \int \frac{|f(x)|^2}{(2\pi\delta a_{\text{eq}}^2)^{1/2}} \exp\left[-\frac{(x + \Delta a_{\bullet})^2}{2\delta a_{\text{eq}}^2}\right] dx, \quad (3)$$

where $\delta a_{\text{eq}}^2 \equiv (\delta A_{\text{eq}})^2/N$ and $\Delta a_{\bullet} = a_{\bullet} - \langle \hat{a} \rangle_{\text{eq}}$. This means that $p(a_{\bullet})$ is a convolution of the distribution in

$\hat{\rho}_{\text{eq}}$ and the shape $|f(x)|^2$ of the measurement operator. We also see that the variance scales as $\delta a_{\bullet}^2 \equiv (\Delta a_{\bullet})^2 = O(1)$, in consistency with $\delta a_{\text{eq}}^2 = O(1)$ and $\delta a_{\text{err}}^2 = O(1)$.

The post-measurement state $\hat{\rho}(a_{\bullet})$ deviates from $\hat{\rho}_{\text{eq}}$, hence the expectation values, denoted by $\langle \cdot \rangle_{a_{\bullet}}$, of observables evolve with time. We investigate the time evolution of an additive observable \hat{B} ($= \sqrt{N}\hat{b}$) for $t > 0$. For $\langle \Delta \hat{b}(t) \rangle_{a_{\bullet}} \equiv \langle \hat{b}(t) \rangle_{a_{\bullet}} - \langle \hat{b} \rangle_{\text{eq}}$, we find [36, 37]

$$\langle \Delta \hat{b}(t) \rangle_{a_{\bullet}} = -\Theta(t) \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} (\ln p)', \quad (4)$$

where $\Theta(t)$ is the step function, $\langle \frac{1}{2} \{ \hat{X}, \hat{Y}(t) \} \rangle_{\text{eq}} \equiv \langle \frac{1}{2} (\hat{X}\hat{Y}(t) + \hat{Y}(t)\hat{X}) \rangle_{\text{eq}}$ the symTC, $\hat{b}(t) = e^{i\hat{H}t/\hbar} \hat{b} e^{-i\hat{H}t/\hbar}$ the Heisenberg operator, and $(\ln p)' = d \ln p / da_{\bullet}$. Since p' vanishes at $\Delta a_{\bullet} = 0$ from Eq. (3), the rhs of Eq. (4) is linear in Δa_{\bullet} for small $|\Delta a_{\bullet}|$.

Averaging Eq. (4) over a_{\bullet} , we find [36]

$$\overline{\langle \Delta \hat{b}(t) \rangle_{a_{\bullet}}} = 0, \text{ i.e., } \overline{\langle \hat{b}(t) \rangle_{a_{\bullet}}} = \langle \hat{b} \rangle_{\text{eq}} \quad (5)$$

for all additive observable $\hat{B} = \sqrt{N}\hat{b}$ at all $t > 0$. That is, the measurement does not cause any systematic disturbance on \hat{b} , in consistency with our requirements on quasiclassical measurements.

With increasing t , $\langle \hat{b}(t) \rangle_{a_{\bullet}}$ relaxes to $\langle \hat{b} \rangle_{\text{eq}}$ if the system possesses ‘mixing property’ in the sense that

$$\lim_{t \rightarrow \infty} \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} = 0, \quad (6)$$

where $\lim_{N \rightarrow \infty}$ is taken before $\lim_{t \rightarrow \infty}$. After the relaxation, one cannot distinguish $\hat{\rho}(a_{\bullet})$ from $\hat{\rho}_{\text{eq}}$ by measuring any additive observables. Equation (4) shows that the relaxation process after the measurement is governed by the symTC. To see it clearly, we calculate the correlation between a_{\bullet} and $\langle \hat{b}(t) \rangle_{a_{\bullet}}$. Multiplying Eq. (4) with Δa_{\bullet} , and averaging over a_{\bullet} , we find [36]

$$\overline{\Delta a_{\bullet} \langle \Delta \hat{b}(t) \rangle_{a_{\bullet}}} = \Theta(t) \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}. \quad (7)$$

This is a universal result independent of choice of $f(x)$, and tells us the operational meaning: *When one measures a time correlation quasiclassically, what he observes is the symTC rather than many other time correlations which reduce to the same classical correlation as $\hbar \rightarrow 0$.*

This result might look contradictory to some experiments [1–3, 38–43]. However, those measurements are not quasiclassical because they destruct the states by absorbing quanta. If, e.g., heterodyning techniques [44, 45] or quantum non-demolition photodetectors [46–50] are used, the symTC will be observed.

Violation of FDT.— The above finding has a great impact on nonequilibrium physics. The Kubo formula [10] gives the response function, which describes response of a quantum system to an external force, by the canonical time correlation, denoted by $\langle \cdot; \cdot \rangle_{\text{eq}}$, of certain additive observables as [23]

$$\Phi_{ba}(t) = \Theta(t) \beta \langle \Delta \hat{a}; \Delta \hat{b}(t) \rangle_{\text{eq}}. \quad (8)$$

Here, the step function $\Theta(t)$ comes from the causality. This formula has been regarded as the FDT for quantum systems [10–12]. In its derivation, however, disturbances by quantum measurements were neglected, although they should be considered seriously [51]. Therefore, we here take Eq. (8) just as a recipe to obtain the response function, as discussed in Ref. [23], while measured fluctuation may possibly be described by a different expression [52]. Then a question arises; does the FDT hold as relations between measured quantities in quantum systems?

To answer this fundamental question, we calculate the Fourier transform of $\Phi_{ba}(t)$, which is the admittance denoted by $\chi_{ba}(\omega)$, and that of the measured time correlation Eq. (7), denoted by $S_{ba}(\omega)$ (whereas the Fourier transform of the symTC without $\Theta(t)$ is denoted by $\tilde{S}_{ba}(\omega)$). We express the results in terms of the symmetric and antisymmetric parts, denoted by $+$ and $-$, respectively, such as $\chi_{ba}^\pm \equiv (\chi_{ba} \pm \chi_{ab})/2$. For the real parts, we have [53]

$$\text{Re } \chi_{ba}^+(\omega) = \beta \text{Re } S_{ba}^+(\omega) / I_\beta(\omega), \quad (9)$$

$$\begin{aligned} \text{Re } \chi_{ba}^-(\omega) &= \beta \text{Re } S_{ba}^-(\omega) \\ &+ \beta \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega' - \omega} \left[1 - \frac{1}{I_\beta(\omega')} \right] i \tilde{S}_{ba}^-(\omega') \frac{d\omega'}{2\pi}. \end{aligned} \quad (10)$$

Here, \mathcal{P} denotes the principal value, and $I_\beta(\omega) \equiv (\beta\hbar\omega/2) \coth(\beta\hbar\omega/2)$. [For the imaginary parts, replace $\text{Re } \chi_{ba}^\pm$, $\text{Re } S_{ba}^\pm$ and $i \tilde{S}_{ba}^\pm$ with $\text{Im } \chi_{ba}^\mp$, $\text{Im } S_{ba}^\mp$ and \tilde{S}_{ba}^\mp , respectively.] Since $I_\beta(\omega) \neq 1$ at finite ω , we find that *the FDT is violated at finite ω , when comparing the observed equilibrium fluctuation and the observed admittance* even if the measurement is quasiclassical (i.e., even if it mimics classical ideal measurement.)

One might expect that the FDT would recover in the ‘classical regime’ where $\hbar\omega \ll k_B T$, because $I_\beta(\omega) \rightarrow 1$. We examine this expectation by studying the case of $\omega = 0$ (for which $\chi_{ba}^\pm(0) = \text{Re } \chi_{ba}^\pm(0)$ because $\Phi_{ba}(t)$ is real). From Eqs. (9) and (10), we find that even at $\omega = 0$ the FDT is recovered only for the symmetric part. For the antisymmetric part, the causality in Eq. (8) (represented by $\Theta(t)$) convolutes different frequencies as

$$\frac{\chi_{ba}^-(0)}{\beta} - S_{ba}^-(0) = \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega} \left[1 - \frac{1}{I_\beta(\omega)} \right] i \tilde{S}_{ba}^-(\omega) \frac{d\omega}{2\pi}. \quad (11)$$

Since $\tilde{S}_{ba}^-(\omega)$ is a pure-imaginary odd function [12], the rhs of Eq. (11) does not vanish in general. Therefore, in general, *the FDT is violated even in the ‘classical regime’ for the antisymmetric part* even if the measurement is quasiclassical. Note that there are two ways to reach the ‘classical regime’ $\beta\hbar\omega \ll 1$. One is to take $\hbar \rightarrow 0$, where the system becomes classical and the violation disappears. The other is to take $\omega \rightarrow 0$ while keeping \hbar constant, where the violation occurs. Therefore, the violation is a genuine quantum effect.

Experiments.— As an example, we compare the conductivity tensor [23], $\sigma_{\mu\nu}(\omega) = \beta \int_0^\infty \langle \hat{j}_\nu; \hat{j}_\mu(t) \rangle_{\text{eq}} e^{i\omega t} dt$,

with the Fourier transform of the measured equilibrium fluctuation, $S_{\mu\nu}(\omega) \equiv \int_0^\infty \langle \frac{1}{2} \{ \hat{j}_\nu, \hat{j}_\mu(t) \} \rangle_{\text{eq}} e^{i\omega t} dt$, where \hat{j}_ν denotes the ν component ($\nu = x, y, z$) of the total current divided by \sqrt{N} .

Koch et al. [44] measured the diagonal ($\mu = \nu$) elements of the equilibrium fluctuation of a circuit by using the heterodyning technique [45]. Their measurement is closer to quasiclassical than those of Refs. [38–43] because it does not destruct states by absorbing quanta, as discussed above. Hence, it can be regarded as a pioneering work about the equilibrium fluctuation obtained by quasiclassical measurements. Its results are consistent with Eq. (9).

For the off-diagonal ($\mu \neq \nu$) elements, no experiments have been reported to the authors’ knowledge. We propose to examine the $\omega = 0$ case, Eq. (11), by studying $\sigma_{xy}(0)$ in the presence of a magnetic field parallel to the z axis. When the system is invariant under rotation by $\pi/2$ about the z axis, the obvious symmetry $\sigma_{xy} = -\sigma_{yx}$ and Eq. (11) yield

$$\frac{\sigma_{xy}(0)}{\beta} - S_{xy}(0) = \int_{-\infty}^{\infty} \frac{\mathcal{P}}{\omega} \left[1 - \frac{1}{I_\beta(\omega)} \right] i \tilde{S}_{xy}^-(\omega) \frac{d\omega}{2\pi}. \quad (12)$$

Therefore, even at $\omega = 0$ the FDT is violated for the off-diagonal (Hall) conductivity. This violation will be confirmed by measuring independently $\sigma_{xy}(0)$ and the correlation $\overline{j_{y\bullet} \langle \hat{j}_x(t) \rangle_{j_{y\bullet}}}$, which gives $S_{xy}(\omega)$ according to Eq. (7). One can also obtain the rhs of Eq. (12) by measuring $\overline{j_{\nu\bullet}^0 j_{\mu\bullet}^k}$ and $\overline{j_{\mu\bullet}^0 j_{\nu\bullet}^k}$, which together give $\tilde{S}_{\mu\nu}(\omega)$ according to Eq. (17) below.

Regression hypothesis.— References [17–19] pointed out that Onsager’s regression hypothesis [13, 14] would contradict with Eq. (8), assuming the symTC for the time correlation of the hypothesis. Nakajima showed that this contradiction can be removed if a local equilibrium state is assumed for the state during fluctuation [15], and thereby derived response to non-mechanical forces [16]. These contradictory claims were derived from different assumptions, but none of them was justified satisfactorily. Since we have proved that the symTC is always measured in quasiclassical experiments, the hypothesis cannot be valid as a relation between measured quantities in quantum systems even if the measurements mimic classical ideal measurements.

Squeezed equilibrium state.— We have shown how quantum effects appear in the expectation value, Eqs. (4) and (5), and in the correlation, Eq. (7). Quantum effects appear more manifestly in the variance of $\hat{b}(t)$, which is calculated as [37]

$$\begin{aligned} & \langle (\hat{b}(t) - \langle \hat{b}(t) \rangle_{a\bullet})^2 \rangle_{a\bullet} - \delta b_{\text{eq}}^2 \\ &= \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} (\ln p)'' - \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}^2 \frac{\partial^2 \ln q}{\partial y^2} \bigg|_{y=0}, \end{aligned} \quad (13)$$

where

$$q(a_\bullet, y) \equiv \langle f(\hat{a} - a_\bullet + y)f(\hat{a} - a_\bullet - y) \rangle_{\text{eq}} \\ = \int \frac{f(x+y)f(x-y)}{(2\pi\delta a_{\text{eq}}^2)^{1/2}} \exp\left[-\frac{(x+\Delta a_\bullet)^2}{2\delta a_{\text{eq}}^2}\right] dx. \quad (14)$$

We find that the relaxation process is governed by both the symTC and the commutator time correlation $\langle \frac{1}{2i}[\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}$. If, in addition to Eq. (6), the system also has the mixing property in the sense that $\lim_{t \rightarrow \infty} \lim_{N \rightarrow \infty} \langle \frac{1}{2i}[\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}^2 = 0$, then the variance relaxes to δb_{eq}^2 with increasing t .

We can see the physical meaning of Eq. (13) by letting $t \rightarrow +0$. The symTC implies that $\hat{\rho}(a_\bullet)$ is squeezed along \hat{b} by measuring \hat{a} if \hat{b} correlates with \hat{a} in $\hat{\rho}_{\text{eq}}$. The commutator time correlation implies that the squeezing is disturbed by the measurement if $[\hat{a}, \hat{b}]$ is non-negligible in $\hat{\rho}_{\text{eq}}$. Furthermore, the rhs of Eq. (4) is $O(1)$, which is the same order as δa_{eq} . That is, $\hat{\rho}(a_\bullet)$ deviates from $\hat{\rho}_{\text{eq}}$ only within equilibrium fluctuations, hence the system remains macroscopically in the same equilibrium state. In this ‘squeezed equilibrium state,’ macrovariables evolve with time as Eq. (4), unlike in the Gibbs state. It represents the state that is observed in quasiclassical measurements of equilibrium fluctuations.

Multi-time measurements.— To measure the lhs’s of Eqs. (4) and (7), one must perform measurements twice in each run of the experiment, as described in Ref. [23]. When one performs more measurements subsequently in each run, what he gets is the following.

Suppose that $K+1$ additive operators $\hat{A}^0, \hat{A}^1, \dots, \hat{A}^K$ are measured subsequently at $t = t_0, t_1, \dots, t_K$, respectively, and the outcomes are $A_\bullet^0, A_\bullet^1, \dots, A_\bullet^K$ ($= \sqrt{N}a_\bullet^0, \sqrt{N}a_\bullet^1, \dots, \sqrt{N}a_\bullet^K$). Here, $K = O(1)$ and $0 = t_0 < t_1 < \dots < t_K$. Let the measurement operator for $\hat{A}^j = \sqrt{N}\hat{a}^j$ be $f_j(\hat{a}^j - a_\bullet^j)$, where $f_j(x)$ satisfies the aforementioned conditions for $f(x)$. The state at $t = t_j$ is obtained by applying $f_j(\hat{a}^j - a_\bullet^j) \dots e^{-i\hat{H}(t_2 - t_1)/\hbar} f_1(\hat{a}^1 - a_\bullet^1) e^{-i\hat{H}t_1/\hbar} f_0(\hat{a}^0 - a_\bullet^0)$ and its conjugate to $\hat{\rho}_{\text{eq}}$ from the left and right, respectively. We find that

$$\overline{\Delta a_\bullet^j} = 0, \text{ i.e., } \overline{a_\bullet^j} = \langle \hat{a}^j \rangle_{\text{eq}} \text{ for all } j. \quad (15)$$

For the correlations for $0 \leq j \leq k \leq K$, we get

$$\overline{\Delta a_\bullet^j \Delta a_\bullet^k} = \langle \frac{1}{2} \{ \Delta \hat{a}^j(t_j), \Delta \hat{a}^k(t_k) \} \rangle_{\text{eq}} + \delta_{j,k} \delta a_{\text{err}}^{j2} \\ + \sum_{l=0}^{j-1} F_l \langle \frac{1}{2i} [\hat{a}^j(t_j), \hat{a}^l(t_l)] \rangle_{\text{eq}} \langle \frac{1}{2i} [\hat{a}^l(t_l), \hat{a}^k(t_k)] \rangle_{\text{eq}}, \quad (16)$$

where $\delta a_{\text{err}}^{j2} = \int x^2 |f_j|^2 dx$ and $F_j = 4 \int \{f'_j(x)\}^2 dx$ [37]. The first term in the rhs of Eq. (16) corresponds to the correlation in Eq. (7). The second term comes from the measurement error of each measurement, which is absent in Eq. (7) because it corresponds to the case $\delta_{j,k} = \delta_{0,1} = 0$. The last term represents disturbances

by the measurements that are performed before the j -th measurement, hence the summation is over m such that $m < j$. Because of this term, which depends on the experimental setup represented by $\{\hat{a}_j, f_j(x), t_j\}_j$, the correlation deviates from the symTC unlike the case of twice measurements, Eq. (7). Consequently, *the FDT is violated more drastically in this protocol of experiment*, e.g., even at $\omega = 0$ both the symmetric and antisymmetric parts can violate the FDT.

For the special case where $j = 0$ (and $k \geq 1$), however, the disturbance term is absent:

$$\overline{\Delta a_\bullet^0 \Delta a_\bullet^k} = \langle \frac{1}{2} \{ \Delta \hat{a}^0, \Delta \hat{a}^k(t_k) \} \rangle_{\text{eq}} \text{ for } t_k > 0. \quad (17)$$

This coincides with Eq. (7), although other measurements may be performed for $0 < t < t_k$. It is also universal, independent of choice of $f(x)$, as is Eq. (7). Hence, in this case, the FDT is violated in the same manner as in the previous protocol.

In summary, we have studied what is measured when the equilibrium fluctuation is measured in an ideal way that mimics classical ideal measurements as closely as possible, i.e., such that disturbances are as small as possible under the condition that the equilibrium fluctuation can be measured accurately enough. We call such measurements quasiclassical. It is found that the symTC is obtained quite generally [Eqs. (7) and (17)]. From this finding and the causality, we have shown that the FDT is violated as a relation between observed quantities even if measurements are quasiclassical [Eqs. (9)–(11)]. It is violated for antisymmetric parts of response functions at all frequencies and for symmetric parts at high frequencies $\hbar\omega \gtrsim k_B T$. Onsager’s regression hypothesis is also violated. These striking results are genuine quantum effects. Disturbances by measurements appear more strongly in the variances of macrovariables [Eq. (13)] and in the case of multi-time measurements [Eq. (16)]. The post-measurement state is shown to be a time-evolving ‘squeezed equilibrium state,’ in which macrovariables fluctuate and relax [Eqs. (4) and (13)]. It represents the state realized during quasiclassical measurements of equilibrium fluctuations.

Finally, we note that our results apply not only to the Gibbs states but also to pure states representing equilibrium states [54–62].

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[1] R. J. Glauber, Phys. Rev. **130**, 2529 (1963).

[2] C. W. Gardiner, *Quantum Noise* (Springer, Berlin, 1991).

- [3] L. Mandel and E. Wolf, *Optical coherence and quantum optics* (Cambridge University Press, 1995).
- [4] K. Koshino and A. Shimizu, *Physics Reports* **412**, 191 (2005), section 4.
- [5] H. M. Wiseman and G. J. Milburn, *Quantum Measurement and Control* (Cambridge University Press, Cambridge, 2010).
- [6] M. A. Nielsen and I. L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, 2000).
- [7] J. B. Jhonson, *Phys. Rev.* **32**, 97 (1928).
- [8] H. Nyquist, *Phys. Rev.* **32**, 110 (1928).
- [9] H. B. Callen and T. A. Welton, *Phys. Rev.* **83**, 34 (1951).
- [10] R. Kubo, *J. Phys. Soc. Jpn.* **12**, 570 (1957).
- [11] R. Kubo, *Rep. Prog. Phys.* **29** 255 (1966).
- [12] R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II: Nonequilibrium Statistical Mechanics* (Springer-Verlag, Berlin, 1985).
- [13] L. Onsager, *Phys. Rev.* **37**, 405 (1931).
- [14] L. Onsager, *Phys. Rev.* **38**, 2265 (1931).
- [15] S. Nakajima, *Busseiron-Kenkyu* **102**, 24 (1956).
- [16] R. Kubo, M. Yokota and S. Nakajima, *J. Phys. Soc. Jpn.* **12**, 1203 (1957).
- [17] R. Kubo and M. Yokota, *Busseiron-Kenkyu* **89**, 99 (1955).
- [18] P. Talkner, *Ann. Phys.* **167**, 390 (1986),
- [19] G. W. Ford and R. F. O'Connell, *Phys. Rev. Lett.* **77**, 798 (1996).
- [20] L. D. Landau and E. M. Lifshitz, *Statistical Physics*, 3rd Edition, Part 1 (Butterworth-Heinemann, Oxford, 1980)
- [21] A. Shimizu and T. Miyadera, *Phys. Rev. Lett.* **89**, 270503 (2002).
- [22] A. Shimizu and T. Morimae, *Phys. Rev. Lett.* **95**, 090401 (2005).
- [23] Supplemental Material following this manuscript.
- [24] D. Goderis and P. Vets, *Commun. Math. Phys.* **122**, 249 (1989).
- [25] T. Matsui, *Rev. Math. Phys.* **14**, 675 (2002).
- [26] V. Jaksic, Y. Pautrat and C.-A. Pillet, *Markov Process. Related Fields* **16**, 59 (2010).
- [27] K. Fujikura, Master Thesis (University of Tokyo, 2015).
- [28] E. H. Lieb and D. W. Robinson, *Commun. Math. Phys.* **28**, 251 (1972).
- [29] B. Nachtergaele and R. Sims, *Theorem. Commun. Math. Phys.* **265**, 119 (2006).
- [30] B. Nachtergaele, H. Raz, B. Schlein, R. Sims, *Commun. Math. Phys.* **286**, 1073 (2009).
- [31] This can be generalized as in Ref. [23].
- [32] T. TerzioĖglu, *Mathematische Annalen*. **182**, 236 (1969).
- [33] L. Hömander, *The Analysis of Linear Partial Differential Operators I: Distribution theory and Fourier Analysis* (2nd ed.). (Springer-Verlag, Berlin, 1990).
- [34] M. Reed and B. Simon, *Methods of Modern Mathematical Physics: Functional Analysis I* (Revised and enlarged ed.). (Academic Press, San Diego, 1980).
- [35] E. M. Stein and R. Shakarchi, *Fourier Analysis: An Introduction (Princeton Lectures in Analysis)*. (Princeton University Press, Princeton, 2003)
- [36] Rigorous derivation is given in Ref. [23].
- [37] As an example, results for the gaussian f are presented in Ref. [23].
- [38] R. Deblock et al., *Science* **301**, 203 (2003).
- [39] P.M. Billangeon et al., *Phys. Rev. Lett.* **96**, 136804 (2006).
- [40] E. Zakka-Bajjani et al., *Phys. Rev. Lett.* **99**, 236803 (2007).
- [41] J. Basset, H. Bouchiat, R. Deblock, *Phys. Rev. Lett.* **105**, 166801 (2010).
- [42] C. Altimiras, O. Parlaveccchio, P. Joyez, D. Vion, P. Roche, D. Esteve, F. Portier, *Phys. Rev. Lett.* **112**, 236803 (2014).
- [43] O. Parlaveccchio, C. Altimiras, J. R. Souquet, P. Simon, I. Safi, P. Joyez, D. Vion, P. Roche, D. Esteve, F. Portier, *Phys. Rev. Lett.* **114**, 126801 (2015).
- [44] R. H. Koch, D. J. Van Harlingen and J. Clarke, *Phys. Rev. B* **26**, 74 (1982).
- [45] G. J. Milburn, *Phys. Rev. A*, **36**, 5271 (1987). In quantum optics the outgoing modes at different times commute with each other. Hence, the last term in Eq. (3.33) of this reference equals the corresponding symTC.
- [46] A. Shimizu, Abstracts of the Spring Meeting of the Physical Society of Japan (Physical Society of Japan, Tokyo, 1990), paper no. 2aR7, in which QND photodetectors using interferometers of massive particles were proposed for the first time.
- [47] A. Shimizu, International Conference on Quantum Electronics: Technical Digest Series 1990 (Optical Society of America, Washington, D.C., 1990), Vol. 8, p. 96.
- [48] A. Shimizu, *Phys. Rev. A* **43**, 3819 (1991).
- [49] M. Brune, S. Haroche, V. Lefevre, J. M. Raimond, and N. Zagury, *Phys. Rev. Lett.* **65**, 976 (1990).
- [50] M. D. Levenson, *Phys. Rev. A* **42**, 2935 (1990).
- [51] H. Takahashi, *J. Phys. Soc. Jpn.* **7**, 439 (1952).
- [52] For other possible problems of the Kubo formula, see, e.g., A. Shimizu and H. Kato, *Nonequilibrium Mesoscopic Conductors Driven by Reservoirs, Low-Dimensional Systems — interactions and Transport Properties* [ed. T. Brandes, Springer, 2000], pp.3-22; arXiv:cond-mat/9911333.
- [53] Although similar relations between χ_{ba} and \tilde{S}_{ba} were known [12], they were not taken seriously because physical significance of the symTC was unknown.
- [54] J. von Neumann, *Z. Phys.* **57**, 30 (1929).
- [55] A. Sugita, *RIMS Kokyuroku (Kyoto)* **1507**, 147 (2006).
- [56] S. Popescu, A.J. Short, and A. Winter, *Nature Phys.* **2**, 754 (2006).
- [57] S. Goldstein, J. L. Lebowitz, R. Tumulka, and N. Zanghi, *Phys. Rev. Lett.* **96**, 050403 (2006).
- [58] A. Sugita, *Nonlinear Phenom. Complex Syst.* **10**, 192 (2007).
- [59] P. Reimann, *Phys. Rev. Lett.* **99**, 160404 (2007).
- [60] S. Sugiura and A. Shimizu, *Phys. Rev. Lett.* **108**, 240401 (2012).
- [61] S. Sugiura and A. Shimizu, *Phys. Rev. Lett.* **111**, 010401 (2013).
- [62] M. Hyuga, S. Sugiura. K. Sakai and A. Shimizu, *Phys. Rev. B* **90**, 121110(R) (2014).

Universal Property of Quantum Measurements of Equilibrium Fluctuations and Violation of Fluctuation-Dissipation Theorem: Supplemental Material

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We first present generalization of measurement operators in Eq. (2). We then give a brief review of the Kubo formula in Sec.II. There, we also discuss irrelevance of disturbances by measurements when measuring response functions. In Sec. III, we explain the experimental protocol for measuring the lhs's of Eqs. (4) and (7). In Sec. IV, we show a version of the conditions and the statement of the quantum central limit theorem (QCLT), which is used for deriving the results of the paper. Furthermore, we present other theorems and their proofs to derive the main results of the paper. In Sec. V, we present derivations of the main results of the paper, showing assumptions on the system and sufficient conditions on $f(x)$. Finally in Sec. VI, we present the results for the case of a gaussian $f(x)$.

I. GENERALIZATION OF MEASUREMENT OPERATORS IN EQ. (2)

We can generalize Eq. (2) as [1]

$$\hat{\rho}(a_{\bullet}) \propto (1/L) \sum_{\lambda=1}^L f^{\lambda}(\hat{a} - a_{\bullet}) \hat{\rho}_{\text{eq}} f^{\lambda}(\hat{a} - a_{\bullet}) \quad (\text{I.1})$$

with L different functions f^1, f^2, \dots, f^L that satisfy the conditions for f of the paper. As long as $L = O(1)$, one can obtain the results for $L > 1$ by averaging the results of the paper over all f^{λ} .

II. BRIEF REVIEW OF KUBO FORMULA

A. Kubo formula

Consider a macroscopic quantum system, which was in an equilibrium state $\hat{\rho}_{\text{eq}} = e^{-\beta \hat{H}}/Z$ in a remote past. Suppose that a spatially-varying time-dependent external field $F(\mathbf{r}, t)$ is applied, which interacts with the system via the interaction Hamiltonian

$$\hat{H}_{\text{ext}}(t) = - \sum_{\mathbf{r}} F(\mathbf{r}, t) \hat{\gamma}(\mathbf{r}), \quad (\text{II.1})$$

where $\hat{\gamma}(\mathbf{r})$ is a ‘local operator’ that acts on a finite number of sites around site \mathbf{r} of the system. Then the state $\hat{\rho}(t)$ of the system deviates from $\hat{\rho}_{\text{eq}}$. Consequently, the expectation value $\langle \alpha(\mathbf{r}) \rangle_t = \text{Tr}[\hat{\rho}(t) \hat{\alpha}(\mathbf{r})]$ of a local observable $\hat{\alpha}(\mathbf{r})$ deviates from its equilibrium value $\langle \alpha(\mathbf{r}) \rangle_{\text{eq}} = \text{Tr}[\hat{\rho}_{\text{eq}} \hat{\alpha}(\mathbf{r})]$. Their difference is the response of $\hat{\alpha}(\mathbf{r})$ to $F(\mathbf{r}, t)$. To the linear order in $F(\mathbf{r}, t)$, the response and $F(\mathbf{r}, t)$ is related by the (linear) *response function* $\phi(\mathbf{r}, t)$ as

$$\langle \alpha(\mathbf{r}) \rangle_t - \langle \alpha(\mathbf{r}) \rangle_{\text{eq}} = \int_{-\infty}^t dt' \sum_{\mathbf{r}'} \phi(\mathbf{r}, \mathbf{r}', t - t') F(\mathbf{r}', t'). \quad (\text{II.2})$$

Although the \mathbf{r} dependence is often crucial,¹ we shall present a simplified case where $F(\mathbf{r}, t)$ is independent of \mathbf{r} ,

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¹ Let \mathbf{k} and ω be the wavenumber and frequency of $F(\mathbf{r}, t)$. In general, when one is interested in the limit of $\mathbf{k} \rightarrow 0$ and $\omega \rightarrow 0$, the two limits should be taken in an appropriate order to get a correct result. This means, for example, that the result for $\mathbf{k} \rightarrow 0$ often disagrees with the result for $\mathbf{k} = 0$, hence the \mathbf{r} dependence cannot be neglected. See Ref. [3] on this point and other issues.

i.e., $F(\mathbf{r}, t) = F(t)$. Then, Eqs. (II.1) and (II.2) are simplified to

$$\hat{H}_{\text{ext}}(t) = -F(t)\hat{C}, \quad (\text{II.3})$$

$$\frac{\langle A \rangle_t}{N} - \frac{\langle A \rangle_{\text{eq}}}{N} = \int_{-\infty}^t \Phi(t-t')F(t')dt', \quad (\text{II.4})$$

where

$$\hat{C} = \sum_{\mathbf{r}} \hat{\gamma}(\mathbf{r}), \quad (\text{II.5})$$

$$\hat{A} = \sum_{\mathbf{r}} \hat{\alpha}(\mathbf{r}) \quad (\text{II.6})$$

are additive observables, N is the number of sites of the system, and

$$\Phi(t) = \frac{1}{N} \sum_{\mathbf{r}} \sum_{\mathbf{r}'} \phi(\mathbf{r}, \mathbf{r}', t) \quad (\text{II.7})$$

is the response function describing the linear response of \hat{A}/N to $F(t)$.

Since we consider the linear response, $F(t)$ is assumed to be small and independent of N . Hence, $\langle A \rangle_t/N$ is also small and independent of N (for sufficiently large N). Therefore, $\Phi(t) = O(1)$ according to Eq. (II.4).

Assuming the unitary time evolution (see Sec. II C) by the total Hamiltonian $\hat{H} + \hat{H}_{\text{ext}}(t)$, Kubo [2] showed that

$$\Phi(t) = \Theta(t) \lim_{N \rightarrow \infty} \frac{1}{i\hbar} \langle [\hat{c}, \hat{a}(t)] \rangle_{\text{eq}}, \quad (\text{II.8})$$

$$= \Theta(t) \lim_{N \rightarrow \infty} \beta \langle \hat{b}; \hat{a}(t) \rangle_{\text{eq}}, \quad (\text{II.9})$$

which is called the Kubo formula. Here, $\hat{c} = \hat{C}/\sqrt{N}$, $\hat{a}(t)$ is the Heisenberg operator ($e^{i\hat{H}t/\hbar} \hat{a} e^{-i\hat{H}t/\hbar}$) of $\hat{a} = \hat{A}/\sqrt{N}$, and $\hat{b} = \hat{B}/\sqrt{N}$, where

$$\hat{B} := \left. \frac{d}{dt} \hat{C}(t) \right|_{t=0} = \frac{1}{i\hbar} [\hat{C}, \hat{H}]. \quad (\text{II.10})$$

If \hat{H} includes only short-range interactions, \hat{B} becomes an additive observable. Furthermore, $\langle ; \rangle_{\text{eq}}$ denotes the *canonical correlation*, which is defined for two operators \hat{X} and \hat{Y} by

$$\langle \hat{X}; \hat{Y} \rangle_{\text{eq}} \equiv \frac{1}{\beta} \int_0^\beta \langle e^{\lambda \hat{H}} \hat{X}^\dagger e^{-\lambda \hat{H}} \hat{Y} \rangle_{\text{eq}} d\lambda. \quad (\text{II.11})$$

Note that

$$\langle \hat{X}; \hat{1} \rangle_{\text{eq}} = \langle \hat{1}; \hat{X} \rangle_{\text{eq}} = \langle \hat{X} \rangle_{\text{eq}}. \quad (\text{II.12})$$

Since

$$\langle \hat{B} \rangle_{\text{eq}} = \left. \frac{d}{dt} \langle \hat{C}(t) \rangle_{\text{eq}} \right|_{t=0} = 0, \quad (\text{II.13})$$

we can rewrite Eq. (II.9), using Eq. (II.12), as

$$\Phi_{ab}(t) = \Theta(t) \lim_{N \rightarrow \infty} \beta \langle \Delta \hat{b}; \Delta \hat{a}(t) \rangle_{\text{eq}}, \quad (\text{II.14})$$

where $\Delta \hat{b} := \hat{b} - \langle \hat{b} \rangle_{\text{eq}}$ ($= \hat{b}$) and $\Delta \hat{a}(t) := \hat{a}(t) - \langle \hat{a} \rangle_{\text{eq}}$, and the subscript ‘ ab ’ of $\Phi_{ab}(t)$ denotes the dependence of the response function on \hat{b} and \hat{a} . This is Eq. (8) of the text (where the roles of \hat{a} and \hat{b} are interchanged).

B. Admittance

The admittance $\chi_{ab}(\omega)$ is the Fourier transform of $\Phi_{ab}(t)$. Its symmetric and antisymmetric parts are defined as in the text by

$$\chi_{ab}^{\pm}(\omega) := \frac{\chi_{ab}(\omega) \pm \chi_{ba}(\omega)}{2}. \quad (\text{II.15})$$

Here, $\chi_{ba}(\omega)$ is the admittance for the case where the roles of \hat{a} and \hat{b} are interchanged. One is often interested in the case where $F(t)$ and \hat{A} are some components of vectors. In such a case, $\Delta\hat{a}$ and $\Delta\hat{b}$ in Eq. (II.14) become the corresponding components of vectors, and the admittance $\chi_{ab}(\omega)$ becomes a tensor, and $\chi_{ab}^{\pm}(\omega)$ its symmetric and antisymmetric tensor components.

For example, one can take $F(t) = E_y(t)$ (the y component of an electric field $\mathbf{E}(t)$) and $\hat{A} = \hat{J}_x$ (the x component of the total current $\hat{\mathbf{J}}$). Then, $\chi_{ab}(\omega)$ is the Hall conductivity $\sigma_{xy}(\omega)$. In particular, when the system is invariant under rotation by $\pi/2$ about the z axis, we also have $\chi_{ab}^{-}(\omega) = \sigma_{xy}(\omega)$ because of the obvious symmetry $\sigma_{xy} = -\sigma_{yx}$.

C. Irrelevance of perturbations by measurements when measuring response functions

The FDT claims that

$$\text{response function} = \beta \times \text{equilibrium fluctuation}. \quad (\text{II.16})$$

Let us consider how accurately the response function and the fluctuation should be measured to confirm the FDT.

To measure the response function, one applies $F(t)$ to the system, which induces a change of \hat{A} of $O(N)$. By measuring this change, one can determine the response function $\Phi(t)$ according to Eq. (II.4). Therefore, for measuring the response function it is sufficient to measure $\langle \hat{A} \rangle_t$ with measurement error of $O(N)$. More concretely, the measurement error for \hat{A} should be $\varepsilon \langle \hat{A} \rangle_{\text{eq}}$, where $0 < \varepsilon \ll 1$.

On the other hand, to measure the equilibrium fluctuation, one does not apply $F(t)$ and watch fluctuations of \hat{A} and \hat{B} . Since their fluctuations are of $O(\sqrt{N})$, one needs to measure \hat{A} and \hat{B} with measurement error of $O(\sqrt{N})$. More concretely, the measurement error for \hat{A} should be $\varepsilon \delta A_{\text{eq}}$, as stated in the text, where $0 < \varepsilon \ll 1$. This measurement error is $O(1/\sqrt{N})$ times smaller than that for the measurement of the response function. This means that *disturbances by measurements affect the fluctuation measurements much more strongly than the measurements of response functions*, because, in general, the (minimum) disturbance by measurement becomes larger as the measurement error is decreased.

In fact, we have shown in our paper that disturbances of quasiclassical measurements are $O(\sqrt{N})$ for additive observables.² For sufficiently large N , such disturbances are negligible for measurements of $\langle \hat{A} \rangle_t/N$ although they play crucial roles in the fluctuation measurements as our paper has shown. Therefore, disturbances by quasiclassical measurements can be neglected in measurements of response functions. For this reason, we have assumed in our paper that the Kubo formula is correct as a recipe to obtain the response function (while measured fluctuation may possibly be described by a different expression), although Kubo [2] calculated the response function without considering disturbances by measurements.

D. Retarded Green function

Among two forms (II.8) and (II.14) of the Kubo formula, formula (II.14) is preferred in statistical mechanics because it resembles the formula for thermodynamic susceptibilities (i.e., response of equilibrium values to equilibrium parameters such as a static magnetic field) for quantum systems. This similarity is convenient, e.g., when a necessary condition for the validity of the Kubo formula is discussed [2, 3].

On the other hand, formula (II.8) is generally preferred in condensed-matter physics because the rhs is nothing but the retarded Green function, which can be calculated using the diagrammatic expansion. In practical calculations, formula (II.8) is often transformed into other forms using, e.g., the charge conservation. For example, the longitudinal conductivity is usually calculated after transforming formula (II.8) into the current-current retarded Green function.

² Although we have shown this fact for equilibrium states, we expect that the order of magnitude does not change in the linear-response regime.

III. PROTOCOL FOR MEASURING THE LHS'S OF EQS. (4) AND (7)

The protocol for measuring the lhs's of Eqs. (4) and (7) is as follows.

One must perform many runs of experiments, which we label $r = 1, 2, \dots$. In each experiment,

- 1) prepare the system in the same equilibrium state,
- 2) measure \hat{a} at time t_r to get the outcome $a_{\bullet r}$,
- 3) measure \hat{b} at time $t_r + t$ to get the outcome $b_{\bullet r}$.

By repeating 1)-3) many times, one gets a set of data $\{a_{\bullet r}, b_{\bullet r}\}_r$. Take arbitrarily a value a_{\bullet} , and select all data that satisfy $|a_{\bullet r} - a_{\bullet}| \ll 1$. By averaging $b_{\bullet r}$ over the selected data, one obtains $\langle \hat{b}(t) \rangle_{a_{\bullet}}$. By subtracting $\langle \hat{b} \rangle_{\text{eq}}$, a way of getting which is obvious, one gets the lhs of Eq. (4). By multiplying this with Δa_{\bullet} , and by averaging over a_{\bullet} , one gets the lhs of Eq. (7).

IV. THEOREMS

We write N for the size, such as the number of spins, of the quantum system. The thermodynamic limit (TDL) is briefly denoted by $\lim_{N \rightarrow \infty}$ throughout this Supplemental Material (and the paper). The justification of interchanging the order of the TDL and differentiation is one of the keys to the rigorous derivations of our results.

We let

$$\hat{A}_i = \sum_{\mathbf{r}} \hat{\alpha}_i(\mathbf{r}) \quad (\text{IV.1})$$

be an additive observable, where $\hat{\alpha}_i(\mathbf{r})$ is a local observable acting around \mathbf{r} , and \mathbf{r} runs all over the volume of the system. The index $i = 1, 2, \dots, M$ labels such additive observables of interest, where $M = O(1)$.

A. Conditions for QCLT

The conditions for the QCLT are summarized as follows.

(C1) The following quantities are $O(1)$ (i.e., they have N -independent upper bounds):

$$\left\| [\hat{A}_i, \hat{\alpha}_j(\mathbf{r})] \right\|, \left\| [\hat{A}_i, [\hat{A}_j, \hat{\alpha}_k(\mathbf{r})]] \right\|. \quad (\text{IV.2})$$

(C2) The following quantity decays faster than $1/|\mathbf{r} - \mathbf{r}'|^{d+\varepsilon}$ ($\varepsilon > 0$ is some constant) with increasing the distance $|\mathbf{r} - \mathbf{r}'|$.

$$\left\langle \frac{1}{2} \{ \Delta \hat{\alpha}_i(\mathbf{r}), \Delta \hat{\alpha}_j(\mathbf{r}') \} \right\rangle_{\text{eq}} \quad (\text{IV.3})$$

(C3) For any simply connected subsystem Λ ,

$$\sum_{\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4 \in \Lambda} |\langle \Delta \hat{\alpha}_i(\mathbf{r}_1) \Delta \hat{\alpha}_j(\mathbf{r}_2) \Delta \hat{\alpha}_k(\mathbf{r}_3) \Delta \hat{\alpha}_l(\mathbf{r}_4) \rangle| = O(|\Lambda|^2). \quad (\text{IV.4})$$

This implies, for example, that the forth moment of any additive operator is $O(N^2)$.

(C4) For some positive constant ε ,

$$N^{1/2} c_N(N^{1/2d-\varepsilon}, N^{1/2d+\varepsilon}; \xi) \rightarrow 0 \quad (N \rightarrow \infty), \quad (\text{IV.5})$$

where

$$\begin{aligned} & c_N(r, N'; \xi) \\ & \equiv \sup \left\{ \left| \langle e^{i\xi_1 \hat{A}_1^{\Lambda+\Lambda'}} / \sqrt{N} \dots e^{i\xi_M \hat{A}_M^{\Lambda+\Lambda'}} / \sqrt{N} \rangle_{\text{eq}} - \langle e^{i\xi_1 \hat{A}_1^{\Lambda}} / \sqrt{N} \dots e^{i\xi_M \hat{A}_M^{\Lambda}} / \sqrt{N} \rangle_{\text{eq}} \langle e^{i\xi_1 \hat{A}_1^{\Lambda'}} / \sqrt{N} \dots e^{i\xi_M \hat{A}_M^{\Lambda'}} / \sqrt{N} \rangle_{\text{eq}} \right|; \right. \\ & \quad \left. d(\Lambda, \Lambda) \geq r, \min\{|\Lambda|, |\Lambda'|\} \leq N' \right\}. \end{aligned} \quad (\text{IV.6})$$

Here, Λ and Λ' are arbitrary simply-connected subsystems, and \hat{A}_j^Λ denotes the restriction of \hat{A}_j to Λ , i.e.,

$$\hat{A}_j^\Lambda = \sum_{\mathbf{r} \in \Lambda} \hat{\alpha}_j(\mathbf{r}). \quad (\text{IV.7})$$

B. Quantum Central Limit Theorems

Theorem 1 (QCLT for the characteristic function). *If the conditions of Sec. IV A are satisfied, then*

$$\lim_{N \rightarrow \infty} \langle \exp[i \frac{\xi_1}{\sqrt{N}} \Delta \hat{A}_1] \cdots \exp[i \frac{\xi_M}{\sqrt{N}} \Delta \hat{A}_M] \rangle_{\text{eq}} = \exp[-\frac{1}{2} \sum_{j,k} \xi_j \xi_k s_{jk}], \quad (\text{IV.8})$$

where

$$s_{jk} \equiv \begin{cases} \lim_{N \rightarrow \infty} \frac{1}{N} \langle \Delta \hat{A}_j \Delta \hat{A}_k \rangle_{\text{eq}} & (j \leq k) \\ \lim_{N \rightarrow \infty} \frac{1}{N} \langle \Delta \hat{A}_k \Delta \hat{A}_j \rangle_{\text{eq}} & (j > k) \end{cases} \quad (\text{IV.9})$$

$$= \begin{cases} \lim_{N \rightarrow \infty} \frac{1}{N} \langle \frac{1}{2} \{ \Delta \hat{A}_j, \Delta \hat{A}_k \} \rangle_{\text{eq}} + \lim_{N \rightarrow \infty} \frac{1}{N} \langle \frac{1}{2} [\Delta \hat{A}_j, \Delta \hat{A}_k] \rangle_{\text{eq}} & (j \leq k) \\ \lim_{N \rightarrow \infty} \frac{1}{N} \langle \frac{1}{2} \{ \Delta \hat{A}_j, \Delta \hat{A}_k \} \rangle_{\text{eq}} + \lim_{N \rightarrow \infty} \frac{1}{N} \langle \frac{1}{2} [\Delta \hat{A}_k, \Delta \hat{A}_j] \rangle_{\text{eq}} & (j > k) \end{cases} \quad (\text{IV.10})$$

This theorem implies that the normalized fluctuation operators $\Delta \hat{A}_j / \sqrt{N}$ behave as c-numbers, i.e. as bosons, in the TDL. Indeed, one can consider that, in the TDL, the fluctuation operators $\Delta \hat{A}_j / \sqrt{N}$ form the CCR (Canonical Commutation Relation) algebra and the state is represented by a gaussian state for the algebra. This theorem is an extension to quantum mechanics of the (classical) central limit theorem, which states that the probabilistic distribution of an additive quantity tends to obey the gaussian distribution. For the detail and proof of the above theorem, see [4–6].

Actually, the conditions of Sec. IV A are a bit stronger than required for the QCLT of this form. The conditions, though, are required for Theorem 3.

Furthermore, we can prove the following QCLTs for the first and second derivatives of the ‘characteristic function,’ which is a quantum analog of the characteristic function. These theorems will be used in Sec. V.

Theorem 2 (QCLT for the first derivative). *If the conditions of Sec. IV A are satisfied, then*

$$\lim_{N \rightarrow \infty} \frac{\partial}{\partial \xi_j} \langle \exp[i \frac{\xi_1}{\sqrt{N}} \Delta \hat{A}_1] \cdots \exp[i \frac{\xi_M}{\sqrt{N}} \Delta \hat{A}_M] \rangle_{\text{eq}} = \frac{\partial}{\partial \xi_j} \lim_{N \rightarrow \infty} \langle \exp[i \frac{\xi_1}{\sqrt{N}} \Delta \hat{A}_1] \cdots \exp[i \frac{\xi_M}{\sqrt{N}} \Delta \hat{A}_M] \rangle_{\text{eq}} \quad (\text{IV.11})$$

$$= \frac{\partial}{\partial \xi_j} \exp \left[-\frac{1}{2} \sum_{k,l} \xi_k \xi_l s_{kl} \right]. \quad (\text{IV.12})$$

Theorem 3 (QCLT for the second derivative). *If the conditions of Sec. IV A are satisfied, then*

$$\lim_{N \rightarrow \infty} \frac{\partial^2}{\partial \xi_j^2} \langle \exp[i \frac{\xi_1}{\sqrt{N}} \Delta \hat{A}_1] \cdots \exp[i \frac{\xi_M}{\sqrt{N}} \Delta \hat{A}_M] \rangle_{\text{eq}} = \frac{\partial^2}{\partial \xi_j^2} \lim_{N \rightarrow \infty} \langle \exp[i \frac{\xi_1}{\sqrt{N}} \Delta \hat{A}_1] \cdots \exp[i \frac{\xi_M}{\sqrt{N}} \Delta \hat{A}_M] \rangle_{\text{eq}} \quad (\text{IV.13})$$

$$= \frac{\partial^2}{\partial \xi_j^2} \exp \left[-\frac{1}{2} \sum_{k,l} \xi_k \xi_l s_{kl} \right]. \quad (\text{IV.14})$$

To prove these theorems, we prepare some lemmas. Hereafter, we call an operator $\hat{U}(\boldsymbol{\xi})$ a *unitary operator generated by fluctuation* if

$$\hat{U}(\boldsymbol{\xi}) = e^{i \xi_1 \Delta \hat{A}_1 / \sqrt{N}} \cdots e^{i \xi_L \Delta \hat{A}_L / \sqrt{N}}, \quad (\text{IV.15})$$

where $\hat{A}_1, \dots, \hat{A}_L$ are additive operators, and L is some constant of $O(1)$. We write $|\boldsymbol{\xi}| = \sum_j |\xi_j|$.

Lemma 1. Assume that the system satisfies condition (C1). Let $\hat{U}(\xi)$ be a unitary operator generated by fluctuation, and \hat{B} an additive operator. Then, there exists a positive constant K of $O(1)$ for which the following inequality holds for any ξ :

$$\frac{1}{\sqrt{N}} \left\| [\hat{U}, \hat{B}] \right\| < K |\xi|. \quad (\text{IV.16})$$

Proof. First, we prove the lemma for the case $\hat{U}(\xi) = e^{i\xi\Delta\hat{A}/\sqrt{N}}$.

$$\begin{aligned} \frac{1}{\sqrt{N}} \left\| [e^{i\xi\Delta\hat{A}/\sqrt{N}}, \hat{B}] \right\| &= \frac{1}{N} \left\| \int_0^\xi d\xi' i e^{i(\xi-\xi')\Delta\hat{A}/\sqrt{N}} [\hat{B}, \hat{A}] e^{i\xi'\Delta\hat{A}/\sqrt{N}} \right\| \\ &\leq |\xi| \frac{1}{N} \left\| [\hat{A}, \hat{B}] \right\|. \end{aligned}$$

By condition (C1) the rhs is $O(1)$. Next, we consider the general case where $\hat{U}(\xi) = e^{i\xi\hat{A}_1/\sqrt{N}} \dots e^{i\xi\hat{A}_L/\sqrt{N}}$.

$$\begin{aligned} \frac{1}{\sqrt{N}} \left\| [e^{i\xi\hat{A}_1/\sqrt{N}} \dots e^{i\xi\hat{A}_L/\sqrt{N}}, \hat{B}] \right\| &\leq \frac{1}{\sqrt{N}} \sum_{j=1}^L \left\| [e^{i\xi\hat{A}_j/\sqrt{N}}, \hat{B}] \right\| \\ &\leq \frac{1}{N} \sum_{j=1}^L |\xi_j| \left\| [\hat{A}_j, \hat{B}] \right\|. \end{aligned}$$

Again, the rhs is $O(1)$ by condition (C1). □

Lemma 2. Assume that the system satisfies condition (C1). Let $\hat{U}(\xi)$ be a unitary operator generated by fluctuation, and \hat{B}_1, \hat{B}_2 be additive operators. Then, there exist positive constants K_1, K_2 of $O(1)$ for which the following inequality holds for any ξ :

$$\frac{1}{N} \left\| [[\hat{U}, \hat{B}_1], \hat{B}_2] \right\| < K_1 |\xi| + K_2 \xi \cdot \xi. \quad (\text{IV.17})$$

Proof. First, we prove the lemma for the case $\hat{U}(\xi) = e^{i\xi\Delta\hat{A}/\sqrt{N}}$.

$$\begin{aligned} &\frac{1}{N} \left\| [[e^{i\xi\Delta\hat{A}/\sqrt{N}}, \hat{B}_1], \hat{B}_2] \right\| \\ &= \frac{1}{N^{3/2}} \left\| \left[\int_0^\xi d\xi' i e^{i(\xi-\xi')\Delta\hat{A}/\sqrt{N}} [\hat{B}_1, \hat{A}] e^{i\xi'\Delta\hat{A}/\sqrt{N}}, \hat{B}_2 \right] \right\| \\ &\leq \frac{1}{N^{3/2}} \left\| \int_0^\xi d\xi' i [e^{i(\xi-\xi')\Delta\hat{A}/\sqrt{N}}, \hat{B}_2] [\hat{B}_1, \hat{A}] e^{i\xi'\Delta\hat{A}/\sqrt{N}} \right\| + \frac{1}{N^{3/2}} \left\| \int_0^\xi d\xi' i e^{i(\xi-\xi')\Delta\hat{A}/\sqrt{N}} [[\hat{B}_1, \hat{A}], \hat{B}_2] e^{i\xi'\Delta\hat{A}/\sqrt{N}} \right\| \\ &\quad + \frac{1}{N^{3/2}} \left\| \int_0^\xi d\xi' i e^{i(\xi-\xi')\Delta\hat{A}/\sqrt{N}} [\hat{B}_1, \hat{A}] [e^{i\xi'\Delta\hat{A}/\sqrt{N}}, \hat{B}_2] \right\| \\ &\leq \frac{1}{N^2} \int_0^\xi d\xi' |\xi - \xi'| \left\| [\hat{A}, \hat{B}_2] \right\| \left\| [\hat{B}_1, \hat{A}] \right\| + \frac{1}{N^{3/2}} \int_0^\xi d\xi' \left\| [[\hat{B}_1, \hat{A}], \hat{B}_2] \right\| + \frac{1}{N^2} \int_0^\xi d\xi' \left\| [\hat{B}_1, \hat{A}] \right\| |\xi'| \left\| [\hat{A}, \hat{B}_2] \right\| \\ &\leq \frac{\xi^2}{2} \frac{1}{N^2} \left\| [\hat{A}, \hat{B}_2] \right\| \left\| [\hat{B}_1, \hat{A}] \right\| + |\xi| \frac{1}{N^{3/2}} \left\| [[\hat{B}_1, \hat{A}], \hat{B}_2] \right\|. \end{aligned}$$

The first term of the rhs is $O(1)$ and the second term is $o(1)$ by condition (C1). Next, we consider the general case

where $\hat{U}(\boldsymbol{\xi}) = e^{i\xi_1 \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_L \hat{A}_L / \sqrt{N}}$.

$$\begin{aligned}
& \frac{1}{N} \left\| [e^{i\xi_1 \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_L \hat{A}_L / \sqrt{N}}, \hat{B}_1], \hat{B}_2 \right\| \\
& \leq \frac{1}{N} \sum_{j=1}^L \left\| [e^{i\xi_1 \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_{j-1} \hat{A}_{j-1} / \sqrt{N}} [e^{i\xi_j \hat{A}_j / \sqrt{N}}, \hat{B}_1] e^{i\xi_{j+1} \hat{A}_{j+1} / \sqrt{N}} \dots e^{i\xi_L \hat{A}_L / \sqrt{N}}, \hat{B}_2] \right\| \\
& \leq \frac{1}{N} \sum_{j=1}^L \left[\left\| [e^{i\xi_1 \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_{j-1} \hat{A}_{j-1} / \sqrt{N}}, \hat{B}_2] \right\| \left\| [e^{i\xi_j \hat{A}_j / \sqrt{N}}, \hat{B}_1] \right\| \right. \\
& \quad \left. + \left\| [e^{i\xi_j \hat{A}_j / \sqrt{N}}, \hat{B}_1], \hat{B}_2 \right\| \right. \\
& \quad \left. + \left\| [e^{i\xi_j \hat{A}_j / \sqrt{N}}, \hat{B}_1] \right\| \left\| [e^{i\xi_{j+1} \hat{A}_{j+1} / \sqrt{N}} \dots e^{i\xi_L \hat{A}_L / \sqrt{N}}, \hat{B}_2] \right\| \right].
\end{aligned}$$

By Lemma 1 and the previous case, Lemma 2 is proved. \square

Lemma 3 (Bound for the first derivative of the ‘characteristic function’). *Assume that the system satisfies conditions (C1) and (C2). Let $\hat{U}_1(\boldsymbol{\xi})$, $\hat{U}_2(\boldsymbol{\xi}')$ be unitary operators generated by fluctuation, and \hat{B} an additive operator. Then, there exist positive constants K_0 and K_1 of $O(1)$ for which the following inequality holds for any $\boldsymbol{\xi}$, $\boldsymbol{\xi}'$:*

$$\frac{1}{\sqrt{N}} |\langle \hat{U}_1(\boldsymbol{\xi}) \Delta \hat{B} \hat{U}_2(\boldsymbol{\xi}') \rangle_{\text{eq}}| < K_0 + K_1 |\boldsymbol{\xi}|. \quad (\text{IV.18})$$

Proof.

$$\begin{aligned}
\frac{1}{\sqrt{N}} |\langle \hat{U}_1(\boldsymbol{\xi}) \Delta \hat{B} \hat{U}_2(\boldsymbol{\xi}') \rangle_{\text{eq}}| & \leq \frac{1}{\sqrt{N}} |\langle \Delta \hat{B} \hat{U}_1(\boldsymbol{\xi}) \hat{U}_2(\boldsymbol{\xi}') \rangle_{\text{eq}}| + \frac{1}{\sqrt{N}} |\langle [\hat{U}_1(\boldsymbol{\xi}), \Delta \hat{B}] \hat{U}_2(\boldsymbol{\xi}') \rangle_{\text{eq}}| \\
& \leq \frac{1}{\sqrt{N}} \langle \Delta \hat{B}^2 \rangle_{\text{eq}}^{1/2} + \frac{1}{\sqrt{N}} \left\| [\hat{U}_1(\boldsymbol{\xi}), \Delta \hat{B}] \right\|.
\end{aligned}$$

The last line follows from the Cauchy-Schwarz inequality. The first term is bounded by a constant of $O(1)$ by condition (C2). Also, the second term is bounded by $|\boldsymbol{\xi}|$ times a constant of $O(1)$ by Lemma 1. \square

Lemma 4 (Bound for the second derivative of the ‘characteristic function’). *Assume that the system satisfies conditions (C1) and (C2). Let $\hat{U}_1(\boldsymbol{\xi}_1)$, $\hat{U}_2(\boldsymbol{\xi}_2)$, $\hat{U}_3(\boldsymbol{\xi}_3)$ be unitary operators generated by fluctuation, and \hat{B}_1 , \hat{B}_2 be additive operators. Then, there exist positive constants K_0 , K_1 and K_2 of $O(1)$ for which the following inequality holds for any $\boldsymbol{\xi}_1$, $\boldsymbol{\xi}_2$, $\boldsymbol{\xi}_3$:*

$$\frac{1}{N} |\langle \hat{U}_1(\boldsymbol{\xi}_1) \Delta \hat{B}_1 \hat{U}_2(\boldsymbol{\xi}_2) \Delta \hat{B}_2 \hat{U}_3(\boldsymbol{\xi}_3) \rangle_{\text{eq}}| \leq K_0 + K_1 (|\boldsymbol{\xi}_1| + |\boldsymbol{\xi}_3|) + K_2 |\boldsymbol{\xi}_1| |\boldsymbol{\xi}_3|. \quad (\text{IV.19})$$

Proof.

$$\begin{aligned}
& \frac{1}{N} |\langle \hat{U}_1(\boldsymbol{\xi}_1) \Delta \hat{B}_1 \hat{U}_2(\boldsymbol{\xi}_2) \Delta \hat{B}_2 \hat{U}_3(\boldsymbol{\xi}_3) \rangle_{\text{eq}}| \\
& \leq \frac{1}{N} |\langle \Delta \hat{B}_1 \hat{U}_1(\boldsymbol{\xi}_1) \hat{U}_2(\boldsymbol{\xi}_2) \hat{U}_3(\boldsymbol{\xi}_3) \Delta \hat{B}_2 \rangle_{\text{eq}}| + \frac{1}{N} |\langle [\hat{U}_1(\boldsymbol{\xi}_1), \Delta \hat{B}_1] \hat{U}_2(\boldsymbol{\xi}_2) \hat{U}_3(\boldsymbol{\xi}_3) \Delta \hat{B}_2 \rangle_{\text{eq}}| \\
& \quad + \frac{1}{N} |\langle \Delta \hat{B}_1 \hat{U}_1(\boldsymbol{\xi}_1) \hat{U}_2(\boldsymbol{\xi}_2) [\Delta \hat{B}_2, \hat{U}_3(\boldsymbol{\xi}_3)] \rangle_{\text{eq}}| + \frac{1}{N} |\langle [\hat{U}_1(\boldsymbol{\xi}_1), \Delta \hat{B}_1] \hat{U}_2(\boldsymbol{\xi}_2) [\Delta \hat{B}_2, \hat{U}_3(\boldsymbol{\xi}_3)] \rangle_{\text{eq}}| \\
& \leq \frac{1}{N} \langle \Delta \hat{B}_1^2 \rangle_{\text{eq}}^{1/2} \langle \Delta \hat{B}_2^2 \rangle_{\text{eq}}^{1/2} + \frac{1}{N} \left\| [\hat{U}_1(\boldsymbol{\xi}_1), \Delta \hat{B}_1] \right\| \langle \Delta \hat{B}_2^2 \rangle_{\text{eq}}^{1/2} \\
& \quad + \frac{1}{N} \langle \Delta \hat{B}_1^2 \rangle_{\text{eq}}^{1/2} \left\| [\Delta \hat{B}_2, \hat{U}_3(\boldsymbol{\xi}_3)] \right\| + \frac{1}{N} \left\| [\hat{U}_1(\boldsymbol{\xi}_1), \Delta \hat{B}_1] \right\| \left\| [\Delta \hat{B}_2, \hat{U}_3(\boldsymbol{\xi}_3)] \right\|.
\end{aligned}$$

The last line follows from the Cauchy-Schwarz inequality. From condition (C2) and Lemma 1, Eq. (IV.19) holds. \square

Lemma 5 (Bound for the third derivative of the ‘characteristic function’). *Assume that the system satisfies conditions (C1), (C2) and (C3). Let $\hat{U}_1(\boldsymbol{\xi}_1)$, $\hat{U}_2(\boldsymbol{\xi}_2)$, $\hat{U}_3(\boldsymbol{\xi}_3)$, $\hat{U}_4(\boldsymbol{\xi}_4)$ be unitary operators generated by fluctuation, and \hat{B}_1 , \hat{B}_2 ,*

\hat{B}_3 be additive operators. Then, there exist positive constants K_0, K_1, K_2 and K_3 of $O(1)$ for which the following inequality holds for any ξ_1, ξ_2, ξ_3 and ξ_4 :

$$\begin{aligned} & \frac{1}{N^{3/2}} \left| \langle \hat{U}_1(\xi_1) \Delta \hat{B}_1 \hat{U}_2(\xi_2) \Delta \hat{B}_2 \hat{U}_3(\xi_3) \Delta \hat{B}_3 \hat{U}_4(\xi_4) \rangle_{\text{eq}} \right| \\ & \leq K_0 + K_1(|\xi_1| + |\xi_2| + |\xi_4|) + K_2(|\xi_1||\xi_3| + |\xi_1||\xi_4| + |\xi_2||\xi_4|) + K_3(|\xi_1| + |\xi_2|)|\xi_1||\xi_4|. \end{aligned} \quad (\text{IV.20})$$

Proof.

$$\begin{aligned} & \frac{1}{N^{3/2}} |\langle \hat{U}_1(\xi_1) \Delta \hat{B}_1 \hat{U}_2(\xi_2) \Delta \hat{B}_2 \hat{U}_3(\xi_3) \Delta \hat{B}_3 \hat{U}_4(\xi_4) \rangle_{\text{eq}}| \\ & \leq \frac{1}{N^{3/2}} |\langle \Delta \hat{B}_1 \Delta \hat{B}_2 \hat{U}_1(\xi_1) \hat{U}_2(\xi_2) \hat{U}_3(\xi_3) \hat{U}_4(\xi_4) \Delta \hat{B}_3 \rangle_{\text{eq}}| + \frac{1}{N^{3/2}} |\langle \Delta \hat{B}_1 [\hat{U}_1(\xi_1) \hat{U}_2(\xi_2), \Delta \hat{B}_2] \hat{U}_3(\xi_3) \hat{U}_4(\xi_4) \Delta \hat{B}_3 \rangle_{\text{eq}}| \\ & \quad + \frac{1}{N^{3/2}} |\langle [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \hat{U}_2(\xi_2) \hat{U}_3(\xi_3) \hat{U}_4(\xi_4) \Delta \hat{B}_2 \Delta \hat{B}_3 \rangle_{\text{eq}}| + \frac{1}{N^{3/2}} |\langle [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \hat{U}_2(\xi_2) [\Delta \hat{B}_2, \hat{U}_3(\xi_3) \hat{U}_4(\xi_4)] \Delta \hat{B}_3 \rangle_{\text{eq}}| \\ & \quad + \frac{1}{N^{3/2}} |\langle \Delta \hat{B}_1 \Delta \hat{B}_2 \hat{U}_1(\xi_1) \hat{U}_2(\xi_2) \hat{U}_3(\xi_3) [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \rangle_{\text{eq}}| + \frac{1}{N^{3/2}} |\langle \Delta \hat{B}_1 [\hat{U}_1(\xi_1) \hat{U}_2(\xi_2), \Delta \hat{B}_2] \hat{U}_3(\xi_3) [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \rangle_{\text{eq}}| \\ & \quad + \frac{1}{N^{3/2}} |\langle \Delta \hat{B}_2 [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \hat{U}_2(\xi_2) \hat{U}_3(\xi_3) [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \rangle_{\text{eq}}| + \frac{1}{N^{3/2}} |\langle [[\hat{U}_1(\xi_1), \Delta \hat{B}_1], \Delta \hat{B}_2] \hat{U}_2(\xi_2) \hat{U}_3(\xi_3) [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \rangle_{\text{eq}}| \\ & \quad + \frac{1}{N^{3/2}} |\langle [\hat{U}_1(\xi_1), \Delta \hat{B}_1] [\hat{U}_2(\xi_2), \Delta \hat{B}_2] \hat{U}_3(\xi_3) [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \rangle_{\text{eq}}| \\ & \leq \frac{1}{N^{3/2}} \langle \Delta \hat{B}_1 \Delta \hat{B}_2 \Delta \hat{B}_1 \rangle_{\text{eq}}^{1/2} \langle \Delta \hat{B}_3^2 \rangle_{\text{eq}}^{1/2} + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1) \hat{U}_2(\xi_2), \Delta \hat{B}_2] \right\| \langle \Delta \hat{B}_1^2 \rangle_{\text{eq}}^{1/2} \langle \Delta \hat{B}_3^2 \rangle_{\text{eq}}^{1/2} \\ & \quad + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \right\| \langle \Delta \hat{B}_3 \Delta \hat{B}_2^2 \Delta \hat{B}_3 \rangle_{\text{eq}}^{1/2} + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \right\| \left\| [\Delta \hat{B}_2, \hat{U}_3(\xi_3) \hat{U}_4(\xi_4)] \right\| \langle \Delta \hat{B}_3^2 \rangle_{\text{eq}}^{1/2} \\ & \quad + \frac{1}{N^{3/2}} \langle \Delta \hat{B}_1 \Delta \hat{B}_2^2 \Delta \hat{B}_1 \rangle_{\text{eq}}^{1/2} \left\| [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \right\| + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1) \hat{U}_2(\xi_2), \Delta \hat{B}_2] \right\| \left\| [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \right\| \langle \Delta \hat{B}_1^2 \rangle_{\text{eq}}^{1/2} \\ & \quad + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \right\| \left\| [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \right\| \langle \Delta \hat{B}_2^2 \rangle_{\text{eq}}^{1/2} + \frac{1}{N^{3/2}} \left\| [[\hat{U}_1(\xi_1), \Delta \hat{B}_1], \Delta \hat{B}_2] \right\| \left\| [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \right\| \\ & \quad + \frac{1}{N^{3/2}} \left\| [\hat{U}_1(\xi_1), \Delta \hat{B}_1] \right\| \left\| [\hat{U}_2(\xi_2), \Delta \hat{B}_2] \right\| \left\| [\Delta \hat{B}_3, \hat{U}_4(\xi_4)] \right\|. \end{aligned}$$

The last line follows from the Cauchy-Schwarz inequality and the definition of the operator norm. By conditions (C2) and (C3) and Lemmas 1 and 2, Eq. (IV.20) is proved. \square

Lemma 6. *Let I be a closed and bounded interval, and $\{g_N(x)\}_N$ be a sequence of twice differentiable functions on I which converges (pointwise) to $g(x)$. Assume that the sequence of second derivatives $\{g_N''(x)\}_N$ are uniformly bounded on I . Then, $\{g_N'(x)\}_N$ converges uniformly to $g'(x)$ (hence we can interchange the differential operation and the limit operation).*

Proof. We use a corollary of the Arzelà-Ascoli theorem : *If a sequence of functions $\{h_N(x)\}_N$ on a closed and bounded interval is uniformly bounded and equicontinuous, and if any subsequences (of the sequence) which converge uniformly have the same limit function $h(x)$ independent of the choice of the subsequence, then the original sequence $\{h_N(x)\}_N$ also converges uniformly to $h(x)$.* Now, by the condition, the sequence of the first derivatives $\{g_N'(x)\}_N$ is uniformly bounded and equicontinuous on I . Additionally, if a subsequence $\{g_{N_k}'(x)\}_k$ ($N_1 < N_2 < \dots$) converges uniformly, its limit is $g'(x)$, independent of $\{N_k\}_k$. Therefore, $\{g_N'(x)\}_N$ converges uniformly to $g'(x)$. \square

Now, we can prove the theorems using these lemmas.

Proof of Theorems 2 and 3. We fix ξ and a closed bounded interval I including ξ . By Lemma 4, the second derivative of $\langle e^{i\xi_1 \Delta \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_1 \Delta \hat{A}_1 / \sqrt{N}} \rangle_{\text{eq}}$ with respect to ξ_i is uniformly bounded on the interval. We can then apply Lemma 6, hence Theorem 2 holds. Similarly, by Lemma 5, the third derivative of $\langle e^{i\xi_1 \Delta \hat{A}_1 / \sqrt{N}} \dots e^{i\xi_1 \Delta \hat{A}_1 / \sqrt{N}} \rangle_{\text{eq}}$ with respect to ξ_i is uniformly bounded on the interval, hence Theorem 3 holds. \square

C. Some more theorems

In this subsection, we present some more theorems which will also be used in the next section for deriving our results. The theorems will be proved under certain conditions on $f(x)$, which will be sufficient conditions for our

results. We also prove that Schwartz functions satisfy all the conditions. As in the main paper, we write

$$\hat{a} = \hat{A}/\sqrt{N}, \quad (\text{IV.21})$$

$$\Delta\hat{a} = \hat{a} - \langle\hat{a}\rangle_{\text{eq}}. \quad (\text{IV.22})$$

Note that a function of a self-adjoint operator, such as $f(\hat{a} - a_\bullet)$, is defined in terms of the spectrum decomposition of the operator.

Theorem 4 (‘characteristic function’ of squeezed equilibrium state). *If the conditions of Sec. IV A are satisfied for $\hat{A}_0, \dots, \hat{A}_M, \hat{B}$, and if f_0, \dots, f_M are bounded piecewise-continuous real functions, then*

$$\begin{aligned} & \lim_{N \rightarrow \infty} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) e^{iu\Delta\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \\ &= \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} f_0[\xi] f_0[\xi'] \cdots f_M[\xi] f_M[\xi'] \\ & \quad \times \lim_{N \rightarrow \infty} \langle e^{i\xi_0(\hat{a}^0 - a_\bullet^0)} \cdots e^{i\xi_M(\hat{a}^M - a_\bullet^M)} e^{iu\Delta\hat{b}} e^{i\xi'_M(\hat{a}^M - a_\bullet^M)} \cdots e^{i\xi'_0(\hat{a}^0 - a_\bullet^0)} \rangle_{\text{eq}} \\ &= \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} f_0[\xi] f_0[\xi'] \cdots f_M[\xi] f_M[\xi'] \\ & \quad \times \exp \left[-i \sum_{k=0}^M (\xi_k + \xi'_k) \Delta a_\bullet^k - \frac{1}{2} \sum_{k,l} (\xi_k + \xi'_k)(\xi_l + \xi'_l) \lim_{N \rightarrow \infty} \langle \frac{1}{2} \{ \Delta\hat{a}^k(t_k), \Delta\hat{a}^l(t_l) \} \rangle_{\text{eq}} \right. \\ & \quad - \frac{1}{2} u^2 \lim_{N \rightarrow \infty} \langle \Delta\hat{b}^2 \rangle_{\text{eq}} - \frac{u}{2} \sum_{k=0}^M \xi_k \lim_{N \rightarrow \infty} \langle \frac{1}{2} \{ \Delta\hat{b}, \Delta\hat{a}^k(t_k) \} \rangle_{\text{eq}} \\ & \quad \left. - \sum_{k < l} (\xi_k - \xi'_k)(\xi_l + \xi'_l) \lim_{N \rightarrow \infty} \langle \frac{1}{2} [\hat{a}^k(t_k), \hat{a}^l(t_l)] \rangle_{\text{eq}} - u \sum_{k=0}^M (\xi_k - \xi'_k) \lim_{N \rightarrow \infty} \langle \frac{1}{2} [\hat{a}^k(t_k), \hat{b}] \rangle_{\text{eq}} \right]. \quad (\text{IV.24}) \end{aligned}$$

Here, $f_k[\xi]$ is the Fourier transform of $f_k(x)$:

$$f_k[\xi] = \int \frac{d\xi}{\sqrt{2\pi}} e^{-i\xi x} f_k(x). \quad (\text{IV.25})$$

Proof. By Theorem 2 in [7], the limit of expectation value in the lhs of Eq. (IV.23) is determined by the QCLT, and the limit can be expressed by the Fourier transform as (IV.24). \square

Theorem 5 (expectation value in squeezed equilibrium state). *Assume that the conditions of Sec. IV A are satisfied for $\hat{A}_0, \dots, \hat{A}_M, \hat{B}$, and that f_0, \dots, f_M are bounded piecewise-continuous real functions. If*

$$\langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta\hat{b}^2 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \leq \text{an upper bound independent of } N, \quad (\text{IV.26})$$

then

$$\begin{aligned} & \lim_{N \rightarrow \infty} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta\hat{b} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \\ &= -i \frac{\partial}{\partial u} \lim_{N \rightarrow \infty} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) e^{iu\Delta\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \Big|_{u=0} \quad (\text{IV.27}) \end{aligned}$$

Proof. By the Cauchy-Schwarz inequality and (IV.26),

$$\begin{aligned} & \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta\hat{b}^2 e^{iu\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right| \\ & \leq \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta\hat{b}^2 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right| \\ & \leq \text{an upper bound independent of } N. \end{aligned}$$

This implies that the second derivative of the ‘characteristic function’

$$\frac{\partial^2}{\partial u^2} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) e^{iu\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \quad (\text{IV.28})$$

is uniformly bounded on any bounded interval of u . Hence, by Theorem 4 and Lemma 6, Eq. (IV.27) is proved. \square

Theorem 6 (variance in the squeezed equilibrium state). *Assume that the conditions of Sec. IV A are satisfied for $\hat{A}_0, \dots, \hat{A}_M, \hat{B}$, and that f_0, \dots, f_M are bounded piecewise-continuous real functions. If*

$$\langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^4 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \leq \text{an upper bound independent of } N, \quad (\text{IV.29})$$

then

$$\begin{aligned} & \lim_{N \rightarrow \infty} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^2 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \\ &= -\frac{\partial^2}{\partial u^2} \lim_{N \rightarrow \infty} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) e^{iu\Delta \hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \Big|_{u=0} \end{aligned} \quad (\text{IV.30})$$

Proof. By the Cauchy-Schwarz inequality and (IV.29),

$$\begin{aligned} & \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^3 e^{iu\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right| \\ & \leq \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^2 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right|^{1/2} \\ & \quad \times \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^4 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right|^{1/2} \\ & \leq \text{an upper bound independent of } N. \end{aligned}$$

This implies that the third derivative of the ‘characteristic function’

$$\frac{\partial^3}{\partial u^3} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) e^{iu\hat{b}} f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \quad (\text{IV.31})$$

is uniformly bounded on any bounded interval of u . Hence, by Theorem 4 and Lemma 6, Eq. (IV.27) is proved. \square

As an example of a class of functions that satisfy the conditions of the above theorems, let us consider a Schwartz function which is defined as follows.

Definition 1 (Schwartz function [8–11]). A *Schwartz function* is a infinitely differentiable function such that for any $n, m = 0, 1, \dots$,

$$\sup_{x \in \mathbb{R}} |x^n f^{(m)}(x)| < \infty. \quad (\text{IV.32})$$

For example, $x^k e^{-x^2}$ is a Schwartz function for any $k > 0$.

A Schwartz function has good properties, such as (i) a Schwartz function is absolutely integrable on \mathbb{R} , (ii) the Fourier transform of a Schwartz function is also a Schwartz function. As a result, Schwartz functions satisfy all the conditions of the above theorems.

Theorem 7. *Assume that the conditions of Sec. IV A are satisfied for $\hat{A}_0, \dots, \hat{A}_M, \hat{B}$. For any Schwartz function f , all the conditions of Theorems 5 and 6 are satisfied.*

Proof. By Lemma 4, there exist positive constants K_0, K_1 and K_2 such that

$$\begin{aligned} & \left| \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_M(\hat{a}^M - a_\bullet^M) \Delta \hat{b}^2 f_M(\hat{a}^M - a_\bullet^M) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \right| \\ & \leq \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} |f_0[\xi] f_0[\xi'] \cdots f_M[\xi] f_M[\xi']| \left| \langle e^{i\xi_0(\hat{a}^0 - a_\bullet^0)} \cdots e^{i\xi_M(\hat{a}^M - a_\bullet^M)} \Delta \hat{b}^2 e^{i\xi'_M(\hat{a}^M - a_\bullet^M)} \cdots e^{i\xi'_0(\hat{a}^0 - a_\bullet^0)} \rangle_{\text{eq}} \right| \\ & \leq \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} |f_0[\xi] f_0[\xi'] \cdots f_M[\xi] f_M[\xi']| \left[K_0 + K_1 \sum_j (|\xi_j| + |\xi'_j|) + K_2 \sum_{j,l} |\xi_j| |\xi_l| \right]. \end{aligned}$$

Since $i\xi_j f_j[\xi_j]$ is the Fourier transform of $f'_j(x)$, it is a Schwartz function. Hence, the integrand above is absolutely integrable, and Eq. (IV.26) is satisfied. Eq. (IV.29) is also proved similarly. \square

Next, we prove theorems for the average value.

Theorem 8. If $f_j \in L^2(\mathbb{R})$ and $\xi|f_j[\xi]|^2$ are absolutely integrable then

$$\begin{aligned} & \lim_{N \rightarrow \infty} \int da_{\bullet}^0 \cdots da_{\bullet}^M \langle f_0(\hat{a}^0 - a_{\bullet}^0) \cdots f_M(\hat{a}^M - a_{\bullet}^M) \Delta \hat{b} f_M(\hat{a}^M - a_{\bullet}^M) \cdots f_0(\hat{a}^M - a_{\bullet}^M) \rangle_{\text{eq}} \\ &= \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}}. \end{aligned} \quad (\text{IV.33})$$

Note that the conditions of this theorem are satisfied if, for example, f_j are continuous and piecewise differentiable and f_j' (on each of their subdomains) are square integrable.

Proof.

$$\begin{aligned} & \int da_{\bullet}^0 \cdots da_{\bullet}^M \langle f_0(\hat{a}^0 - a_{\bullet}^0) \cdots f_M(\hat{a}^M - a_{\bullet}^M) \Delta \hat{b} f_M(\hat{a}^M - a_{\bullet}^M) \cdots f_0(\hat{a}^M - a_{\bullet}^M) \rangle_{\text{eq}} \\ &= \int da_{\bullet}^0 \cdots da_{\bullet}^M \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} f_0[\xi_0] f_0^*[\xi'_0] \cdots f_M[\xi_M] f_M^*[\xi'_M] \\ & \quad \times \langle e^{i\xi_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \cdots e^{i\xi_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \Delta \hat{b} e^{-i\xi'_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \cdots e^{-i\xi'_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \rangle_{\text{eq}} \\ &= \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}}. \end{aligned}$$

By Lemma 3, there are $O(1)$ -positive constants K_0 and K_1 , and the integrand of the last integral is bounded by

$$\begin{aligned} & |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \left| \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \right| \\ & \leq |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| \right). \end{aligned}$$

By assumption and the fact that the Fourier transform of any $L^2(\mathbb{R})$ function is also an $L^2(\mathbb{R})$ function, the above upper bound is absolutely integrable:

$$\int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| \right) < \infty.$$

Hence, by the dominated convergence theorem,

$$\begin{aligned} & \lim_{N \rightarrow \infty} \int da_{\bullet}^0 \cdots da_{\bullet}^M \langle f_0(\hat{a}^0 - a_{\bullet}^0) \cdots f_M(\hat{a}^M - a_{\bullet}^M) \Delta \hat{b} f_M(\hat{a}^M - a_{\bullet}^M) \cdots f_0(\hat{a}^M - a_{\bullet}^M) \rangle_{\text{eq}} \\ &= \lim_{N \rightarrow \infty} \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\ &= \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}}. \end{aligned}$$

□

Theorem 9. If $f_j \in L^2(\mathbb{R})$ and $f_j[\xi]$ are continuous and piecewise differentiable, and if $\xi f_j'[\xi] f_j[\xi]$ and $\xi^2 |f_j[\xi]|^2$ are absolutely integrable, then

$$\begin{aligned} & \lim_{N \rightarrow \infty} \int da_{\bullet}^0 \cdots da_{\bullet}^M \Delta a_{\bullet}^l \langle f_0(\hat{a}^0 - a_{\bullet}^0) \cdots f_M(\hat{a}^M - a_{\bullet}^M) \Delta \hat{b} f_M(\hat{a}^M - a_{\bullet}^M) \cdots f_0(\hat{a}^M - a_{\bullet}^M) \rangle_{\text{eq}} \\ &= \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_{l-1}[\xi_{l-1}]|^2 \text{Im}(f_l'[\xi_l] f_l^*[\xi_l]) |f_{l+1}[\xi_{l+1}]|^2 \cdots |f_M[\xi_M]|^2 \\ & \quad \times \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\ & \quad - i \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \\ & \quad \times \lim_{N \rightarrow \infty} \frac{\partial}{\partial s} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{l-1} \Delta \hat{a}^{l-1}} e^{i(\xi_l + s/2) \Delta \hat{a}^l} e^{i\xi_{l+1} \Delta \hat{a}^{l+1}} \cdots e^{i\xi_M \Delta \hat{a}^M} \\ & \quad \times \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_{l+1} \Delta \hat{a}^{l+1}} e^{-i(\xi_l + s/2) \Delta \hat{a}^l} e^{-i\xi_{l-1} \Delta \hat{a}^{l-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \Big|_{s=0}. \end{aligned}$$

Note that the conditions of this theorem are satisfied if, for example, f_j are continuously differentiable and piecewise twice differentiable, and f_j, xf_j, f'_j, f''_j (on each of their subdomains) are square integrable.

Proof.

$$\begin{aligned}
& \int da_{\bullet}^0 \cdots da_{\bullet}^M \Delta a_{\bullet}^l \langle f_0(\hat{a}^0 - a_{\bullet}^0) \cdots f_M(\hat{a}^M - a_{\bullet}^M) \Delta \hat{b} f_M(\hat{a}^M - a_{\bullet}^M) \cdots f_0(\hat{a}^M - a_{\bullet}^M) \rangle_{\text{eq}} \\
&= \int da_{\bullet}^0 \cdots da_{\bullet}^M \Delta a_{\bullet}^l \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} f_0[\xi_0] f_0^*[\xi'_0] \cdots f_M[\xi_M] f_M^*[\xi'_M] \\
&\quad \times \langle e^{i\xi_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \cdots e^{i\xi_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \Delta \hat{b} e^{-i\xi'_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \cdots e^{-i\xi'_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \rangle_{\text{eq}} \\
&= i \int da_{\bullet}^0 \cdots da_{\bullet}^M \int \frac{d\xi_0 d\xi'_0}{2\pi} \cdots \frac{d\xi_M d\xi'_M}{2\pi} \frac{\partial}{\partial(\xi_l - \xi'_l)} \left\{ f_0[\xi_0] f_0^*[\xi'_0] \cdots f_M[\xi_M] f_M^*[\xi'_M] \right. \\
&\quad \times \langle e^{i\xi_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \cdots e^{i\xi_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \Delta \hat{b} e^{-i\xi'_M(\Delta \hat{a}^M - \Delta a_{\bullet}^M)} \cdots e^{-i\xi'_0(\Delta \hat{a}^0 - \Delta a_{\bullet}^0)} \rangle_{\text{eq}} \left. \right\} \\
&= -i \frac{\partial}{\partial s} \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_{l-1}[\xi_{l-1}]|^2 f_l[\xi_l + \frac{s}{2}] f_l^*[\xi_l - \frac{s}{2}] |f_{l+1}[\xi_{l+1}]|^2 \cdots |f_M[\xi_M]|^2 \\
&\quad \times \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{l-1} \Delta \hat{a}^{l-1}} e^{i(\xi_l + s/2) \Delta \hat{a}^l} e^{i\xi_{l+1} \Delta \hat{a}^{l+1}} \cdots e^{i\xi_M \Delta \hat{a}^M} \\
&\quad \times \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_{l+1} \Delta \hat{a}^{l+1}} e^{-i(\xi_l - s/2) \Delta \hat{a}^l} e^{-i\xi_{l-1} \Delta \hat{a}^{l-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \Big|_{s=0} \\
&= \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_{l-1}[\xi_{l-1}]|^2 \text{Im}(f'_l[\xi_l] f_l^*[\xi_l]) |f_{l+1}[\xi_{l+1}]|^2 \cdots |f_M[\xi_M]|^2 \\
&\quad \times \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_M \Delta \hat{a}^M} \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\
&\quad - i \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \\
&\quad \times \frac{\partial}{\partial s} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{l-1} \Delta \hat{a}^{l-1}} e^{i(\xi_l + s/2) \Delta \hat{a}^l} e^{i\xi_{l+1} \Delta \hat{a}^{l+1}} \cdots e^{i\xi_M \Delta \hat{a}^M} \\
&\quad \times \Delta \hat{b} e^{-i\xi_M \Delta \hat{a}^M} \cdots e^{-i\xi_{l+1} \Delta \hat{a}^{l+1}} e^{-i(\xi_l + s/2) \Delta \hat{a}^l} e^{-i\xi_{l-1} \Delta \hat{a}^{l-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \Big|_{s=0}.
\end{aligned}$$

By Lemmas 3 and 4, there are $O(1)$ -positive constants K_0 , K_1 and K_2 , and the integrand above is bounded by

$$\begin{aligned}
& |f_0[\xi_0]|^2 \cdots |f_{l-1}[\xi_{l-1}]|^2 |\text{Im}(f'_l[\xi_l] f_l^*[\xi_l])| |f_{l+1}[\xi_{l+1}]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| \right) \\
& + |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| + K_2 \left(\sum_{j=0}^M |\xi_j| \right)^2 \right).
\end{aligned}$$

By assumption, the above upper bound is absolutely integrable:

$$\begin{aligned}
& \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_{l-1}[\xi_{l-1}]|^2 |\text{Im}(f'_l[\xi_l] f_l^*[\xi_l])| |f_{l+1}[\xi_{l+1}]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| \right) \\
& + \int d\xi_0 \cdots d\xi_M |f_0[\xi_0]|^2 \cdots |f_M[\xi_M]|^2 \left(K_0 + K_1 \sum_{j=0}^M |\xi_j| + K_2 \left(\sum_{j=0}^M |\xi_j| \right)^2 \right) < \infty.
\end{aligned}$$

Hence, by the dominated convergence theorem, the theorem is proved. \square

V. DERIVATIONS OF THE MAIN RESULTS

We now derive the main results of the paper using the above theorems.

A. Assumption on macroscopic systems and conditions on f

We assume that conditions (C1)-(C4) of Sec. IV A are satisfied for all additive observables of interest, $\hat{A}_1, \hat{A}_2, \dots, \hat{A}_M$, where $M = O(1)$. We assume this not only for equal-time operators $\hat{A}_1(t), \hat{A}_2(t), \dots, \hat{A}_M(t)$, but also for operators at different times $\hat{A}_1(t_1), \hat{A}_2(t_2), \dots, \hat{A}_M(t_M)$, where t_1, t_2, \dots, t_M denote the times of measurements, all of which are finite and independent of N .

Let us discuss this assumption for the case where the system is composed of several units, each with a finite dimensional Hilbert space. For equal-time operators, the above assumption is believed to hold generally except in special regions, such as critical points, in the thermodynamic configuration space. For different-time operators, their correlations are bounded by the Lieb-Robinson bounds [12, 13] and therefore by the equal-time correlations, if the interaction is finite range. Hence, the above assumption holds for many quantum systems except in special regions in the thermodynamic configuration space if the interaction is finite range and if the system is composed of several units, each with a finite dimensional Hilbert space.

For the case where each unit has an infinite-dimensional Hilbert space, some of additive observables of interest might have infinite norms, for which condition (C1) is not satisfied. We have not yet succeeded in weakening condition (C1) (while the Lieb-Robinson bounds have been extended to certain continuous quantum systems [14]). Therefore, at present, it is necessary to introduce a certain cutoff, by which each unit is well described by a finite-dimensional Hilbert space. If such an effective theory can be constructed conditions (C1)-(C4) can be satisfied.

For the conditions on f , we will describe a *sufficient* condition on f for *each* result. Hence, a sufficient condition on f for *all* the results is the logical conjunction of these conditions. We think the conditions are general enough from a physical viewpoint, although it might be possible to prove the results under weaker conditions on f .

Note that Schwartz functions satisfy all the conditions on f , as is easily seen from their general properties and Theorem 7.

B. Derivation of Eq. (3)

A sufficient condition on f for Eq. (3) is that f is a bounded piecewise-continuous real function.

By the QCLT, the probability distribution of fluctuation of any additive quantity which satisfies the conditions in Sec. IV A converges weakly to the normal distribution in the TDL. Therefore, for any bounded piecewise-continuous real function f ,

$$\begin{aligned} p(a_\bullet) &\equiv \lim_{N \rightarrow \infty} \langle \{f(\hat{a} - a_\bullet)\}^2 \rangle_{\text{eq}} \\ &= \int \frac{|f(a - a_\bullet)|^2}{(2\pi\delta a_{\text{eq}}^2)^{1/2}} \exp \left[-\frac{(a - \langle \hat{a} \rangle_{\text{eq}})^2}{2\delta a_{\text{eq}}^2} \right] da \\ &= \int \frac{|f(x)|^2}{(2\pi\delta a_{\text{eq}}^2)^{1/2}} \exp \left[-\frac{(x + \Delta a_\bullet)^2}{2\delta a_{\text{eq}}^2} \right] dx. \end{aligned} \quad (\text{V.1})$$

For example, in the case where f is gaussian $f(x) = (2\pi w^2)^{-1/4} \exp(-x^2/4w^2)$,

$$p(a_\bullet) = \frac{1}{\sqrt{2\pi(w^2 + \delta a_{\text{eq}}^2)}} \exp \left[-\frac{1}{2} \frac{\Delta a_\bullet^2}{w^2 + \delta a_{\text{eq}}^2} \right]. \quad (\text{V.2})$$

C. Derivation of Eq. (4) and Eq. (13)

A sufficient condition on f for Eqs. (4) and (13) is that f is a bounded piecewise-continuous real function and that conditions (IV.26) and (IV.29) are satisfied.

To derive Eq. (4) and Eq. (13), we calculate the ‘characteristic function’ of $\Delta \hat{b}(t)$ for the state $\hat{\rho}(a_\bullet)$:

$$\langle \exp(iu\Delta \hat{b}(t)) \rangle_{a_\bullet} = \frac{1}{p(a_\bullet)} \langle f(\hat{a} - a_\bullet) \exp(iu\Delta \hat{b}(t)) f(\hat{a} - a_\bullet) \rangle_{\text{eq}}, \quad (\text{V.3})$$

where u is a real parameter. The expectation value and the variance of $\hat{b}(t)$ are obtained, respectively, by

$$\langle \hat{b}(t) \rangle_{a_\bullet} = -i \frac{\partial}{\partial u} \langle \exp(iu \Delta \hat{b}(t)) \rangle_{a_\bullet} \Big|_{u=0}, \quad (\text{V.4})$$

$$\langle (\hat{b}(t) - \langle \hat{b}(t) \rangle_{a_\bullet})^2 \rangle_{a_\bullet} = -\frac{\partial^2}{\partial u^2} \ln \langle \exp(iu \Delta \hat{b}(t)) \rangle_{a_\bullet} \Big|_{u=0}. \quad (\text{V.5})$$

Furthermore, by Theorems 5 and 6, we can interchange the limit and differentiation. Hence, by Theorem 4,

$$\begin{aligned} & \langle \exp(iu \Delta \hat{b}(t)) \rangle_{a_\bullet} \\ &= \frac{1}{p(a_\bullet)} \frac{1}{2\pi} \int d\xi d\xi' f[\xi] f[\xi'] \langle e^{i\xi(\hat{a}-a_\bullet)} e^{iu \Delta \hat{b}(t)} e^{i\xi'(\hat{a}-a_\bullet)} \rangle_{\text{eq}} \\ & \xrightarrow{N \rightarrow \infty} \frac{1}{p(a_\bullet)} \frac{1}{2\pi} \int d\xi d\xi' f[\xi] f[\xi'] \exp \left[-i(\xi + \xi') \Delta a_\bullet - \frac{1}{2}(\xi + \xi')^2 \delta a_{\text{eq}}^2 \right. \\ & \quad \left. - (\xi + \xi') u \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} - \frac{1}{2} u^2 \delta b(t)_{\text{eq}}^2 \right. \\ & \quad \left. - (\xi - \xi') u \langle \frac{1}{2} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}} \right] \\ &= \frac{1}{p(a_\bullet)} \frac{1}{\sqrt{2\pi \delta a_{\text{eq}}^2}} e^{-u^2 \delta b(t)_{\text{eq}}^2 / 2} \int dx f(x - u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}) f(x + u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}) \\ & \quad \times \exp \left[-\frac{1}{2\delta a_{\text{eq}}^2} (x + \Delta a_\bullet - iu \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}})^2 \right] \\ &\equiv \frac{1}{p(a_\bullet)} e^{-u^2 \delta b(t)_{\text{eq}}^2 / 2} q(a_\bullet - iu \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}, u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}). \end{aligned} \quad (\text{V.6})$$

$$\equiv \frac{1}{p(a_\bullet)} e^{-u^2 \delta b(t)_{\text{eq}}^2 / 2} q(a_\bullet - iu \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}, u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}). \quad (\text{V.7})$$

In the last line, we have defined the following function:

$$q(a'_\bullet, y) \equiv \frac{1}{\sqrt{2\pi \delta a_{\text{eq}}^2}} \int dx f(x - y) f(x + y) \exp \left[-\frac{1}{2\delta a_{\text{eq}}^2} (x + \Delta a'_\bullet)^2 \right]. \quad (\text{V.8})$$

Note that $q(a'_\bullet, 0) = p(a'_\bullet)$ and $(\partial/\partial y)q(a'_\bullet, y)|_{y=0} = 0$.

By using these relations, we have

$$\begin{aligned} \langle \hat{b}(t) \rangle_{a_\bullet} &= -i \frac{\partial}{\partial u} \ln \langle \exp(iu \Delta \hat{b}(t)) \rangle_{a_\bullet} \Big|_{u=0} \\ & \xrightarrow{N \rightarrow \infty} -i \frac{\partial}{\partial u} \ln q(a_\bullet - iu \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}, u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}) \Big|_{u=0} \\ &= -\langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} \frac{\partial}{\partial a_\bullet} \ln q(a_\bullet, 0) - i \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}} \frac{\partial}{\partial y} \ln q(a_\bullet, y) \Big|_{y=0} \\ &= -\langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} (\ln p)'. \end{aligned} \quad (\text{V.9})$$

Similarly,

$$\begin{aligned} & \langle (\hat{b}(t) - \langle \hat{b}(t) \rangle_{a_\bullet})^2 \rangle_{a_\bullet} \\ &= -\frac{\partial^2}{\partial u^2} \ln \langle \exp(iu \Delta \hat{b}(t)) \rangle_{a_\bullet} \Big|_{u=0} \\ & \xrightarrow{N \rightarrow \infty} \delta b_{\text{eq}}^2 - \frac{\partial^2}{\partial u^2} \ln q(a_\bullet - iu \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}, u \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}) \Big|_{u=0} \\ &= \delta b_{\text{eq}}^2 + \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}^2 \frac{\partial^2}{\partial a_\bullet^2} \ln q(a_\bullet, 0) \\ & \quad + i \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}} \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}} \frac{\partial}{\partial a_\bullet} \frac{\partial}{\partial y} \ln q(a_\bullet, y) \Big|_{y=0} - \langle \frac{1}{2i} [\Delta \hat{a}, \Delta \hat{b}(t)] \rangle_{\text{eq}}^2 \frac{\partial^2}{\partial y^2} \ln q(a_\bullet, y) \Big|_{y=0} \\ &= \delta b_{\text{eq}}^2 + \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}^2 \frac{\partial^2}{\partial a_\bullet^2} \ln p(a_\bullet) - \langle \frac{1}{2i} [\Delta \hat{a}, \Delta \hat{b}(t)] \rangle_{\text{eq}}^2 \frac{\partial^2}{\partial y^2} \ln q(a_\bullet, y) \Big|_{y=0}. \end{aligned} \quad (\text{V.10})$$

D. Derivation of Eqs. (5) and (15)

Since Eq. (5) can be obtained as a special case of Eq. (15), we prove the latter.

A sufficient condition on f_j for Eq. (15) is that $f_j \in L^2(\mathbb{R})$ and $\xi|f_j[\xi]|^2$ are absolutely integrable. This condition is satisfied if, for example, f_j are continuous and piecewise differentiable and f'_j (on each of their subdomains) are square integrable.

By Theorem 8, and $\int dx |f_j(x)|^2 = 1$ and $\int dx x|f_j(x)|^2 = 0$, and the spectrum decomposition of $f_j(\hat{a}^j - a_\bullet^j)$,

$$\overline{\Delta a_\bullet^j} \equiv \int da_\bullet^0 da_\bullet^1 \cdots da_\bullet^j \Delta a_\bullet^j \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_j(\hat{a}^j - a_\bullet^j) f_j(\hat{a}^j - a_\bullet^j) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \quad (\text{V.11})$$

$$= \int da_\bullet^0 da_\bullet^1 \cdots da_\bullet^{j-1} dx \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_{j-1}(\hat{a}^{j-1} - a_\bullet^{j-1}) f_j(x) \times (\Delta \hat{a}^j - x) f_j(x) f_{j-1}(\hat{a}^{j-1} - a_\bullet^{j-1}) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \quad (\text{V.12})$$

$$= \int da_\bullet^0 da_\bullet^1 \cdots da_\bullet^{j-1} \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_{j-1}(\hat{a}^{j-1} - a_\bullet^{j-1}) \Delta \hat{a}^j f_{j-1}(\hat{a}^{j-1} - a_\bullet^{j-1}) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \\ \xrightarrow{N \rightarrow \infty} \int d\xi_0 \cdots d\xi_{j-1} |f_0[\xi_0]|^2 \cdots |f_{j-1}[\xi_{j-1}]|^2 \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{j-1} \Delta \hat{a}^{j-1}} \Delta \hat{a}^j e^{-i\xi_{j-1} \Delta \hat{a}^{j-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\ = \int d\xi_0 \cdots d\xi_{j-1} |f_0[\xi_0]|^2 \cdots |f_{j-1}[\xi_{j-1}]|^2 i \sum_{l=0}^{j-1} \xi_l \langle [\hat{a}^l, \hat{a}^j] \rangle_{\text{eq}} \\ = 0. \quad (\text{V.13})$$

E. Derivation of Eq. (7) and Eq. (16)

A sufficient condition on f for Eqs. (7) and (16) is that $f_j \in L^2(\mathbb{R})$ and $f_j[\xi]$ are continuous and piecewise differentiable, and that $\xi f'_j[\xi] f_j[\xi]$ and $\xi^2 |f_j[\xi]|^2$ are absolutely integrable. These conditions are satisfied if, for example, f_j are continuously differentiable and piecewise twice differentiable, and $f_j, x f_j, f'_j, f''_j$ (on each of their subdomains) are square integrable.

We first prove Eq. (16). By Theorem 9, we have for $j < k$

$$\overline{\Delta a_\bullet^j \Delta a_\bullet^k} \equiv \int da_\bullet^0 da_\bullet^1 \cdots da_\bullet^k \Delta a_\bullet^j \Delta a_\bullet^k \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_k(\hat{a}^k - a_\bullet^k) f_k(\hat{a}^k - a_\bullet^k) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \quad (\text{V.14})$$

$$= \int da_\bullet^0 da_\bullet^1 \cdots da_\bullet^{k-1} \Delta a_\bullet^j \langle f_0(\hat{a}^0 - a_\bullet^0) \cdots f_{k-1}(\hat{a}^{k-1} - a_\bullet^{k-1}) \Delta \hat{a}^k f_{k-1}(\hat{a}^{k-1} - a_\bullet^{k-1}) \cdots f_0(\hat{a}^M - a_\bullet^M) \rangle_{\text{eq}} \\ \xrightarrow{N \rightarrow \infty} \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{j-1}[\xi_{j-1}]|^2 \text{Im}(f'_j[\xi_j] f_j^*[\xi_j]) |f_{j+1}[\xi_{j+1}]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \\ \times \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{k-1} \Delta \hat{a}^{k-1}} \Delta \hat{a}^k e^{-i\xi_{k-1} \Delta \hat{a}^{k-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\ + \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \\ \times \lim_{N \rightarrow \infty} \text{Re} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{j-1} \Delta \hat{a}^{j-1}} e^{i\xi_j \Delta \hat{a}^j} \Delta \hat{a}^j e^{i\xi_{j+1} \Delta \hat{a}^{j+1}} \cdots e^{i\xi_{k-1} \Delta \hat{a}^{k-1}} \\ \times \Delta \hat{a}^k e^{-i\xi_{k-1} \Delta \hat{a}^{k-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}}. \quad (\text{V.15})$$

By Theorem 3, we can evaluate these limits by the QCLT as

$$\begin{aligned}
& \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{j-1}[\xi_{j-1}]|^2 \operatorname{Im}(f'_j[\xi_j] f_j^*[\xi_j]) |f_{j+1}[\xi_{j+1}]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \\
& \quad \lim_{N \rightarrow \infty} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{k-1} \Delta \hat{a}^{k-1}} \Delta \hat{a}^k e^{-i\xi_{k-1} \Delta \hat{a}^{k-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle_{\text{eq}} \\
& = - \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{j-1}[\xi_{j-1}]|^2 \operatorname{Im}(f'_j[\xi_j] f_j^*[\xi_j]) |f_{j+1}[\xi_{j+1}]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \sum_{l=0}^{k-1} \xi_l \langle \frac{1}{i} [\hat{a}^l, \hat{a}^k] \rangle_{\text{eq}} \\
& = - \int d\xi_j \operatorname{Im}(f'_j[\xi_j] f_j^*[\xi_j]) \xi_j \langle \frac{1}{i} [\hat{a}^j, \hat{a}^k] \rangle_{\text{eq}} \\
& = \langle \frac{1}{i} [\hat{a}^j, \hat{a}^k] \rangle_{\text{eq}} \operatorname{Im} \int dx x f_j(x) f'_j(x) \\
& = 0,
\end{aligned} \tag{V.16}$$

$$\begin{aligned}
& \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \lim_{N \rightarrow \infty} \operatorname{Re} \langle e^{i\xi_0 \Delta \hat{a}^0} \cdots e^{i\xi_{j-1} \Delta \hat{a}^{j-1}} e^{i\xi_j \Delta \hat{a}^j} \Delta \hat{a}^j e^{i\xi_{j+1} \Delta \hat{a}^{j+1}} \cdots e^{i\xi_{k-1} \Delta \hat{a}^{k-1}} \\
& \quad \Delta \hat{a}^k e^{-i\xi_{k-1} \Delta \hat{a}^{k-1}} \cdots e^{-i\xi_0 \Delta \hat{a}^0} \rangle \\
& = \int d\xi_0 \cdots d\xi_{k-1} |f_0[\xi_0]|^2 \cdots |f_{k-1}[\xi_{k-1}]|^2 \left(\langle \frac{1}{2} \{ \hat{a}^j, \hat{a}^k \} \rangle_{\text{eq}} + \sum_{l=0}^{k-1} \xi_l \langle \frac{1}{i} [\hat{a}^l, \hat{a}^k] \rangle_{\text{eq}} \sum_{h=0}^{j-1} \xi_h \langle \frac{1}{i} [\hat{a}^h, \hat{a}^j] \rangle_{\text{eq}} \right) \\
& = \langle \frac{1}{2} \{ \hat{a}^j, \hat{a}^k \} \rangle_{\text{eq}} + \sum_{l=0}^{j-1} \langle \frac{1}{i} [\hat{a}^l, \hat{a}^k] \rangle_{\text{eq}}^2 \int d\xi_l \xi_l^2 |f_l[\xi_l]|^2 \\
& = \langle \frac{1}{2} \{ \hat{a}^j, \hat{a}^k \} \rangle_{\text{eq}} + \sum_{l=0}^{j-1} F_l \langle \frac{1}{2i} [\hat{a}^j, \hat{a}^l] \rangle_{\text{eq}} \langle \frac{1}{2i} [\hat{a}^l, \hat{a}^k] \rangle_{\text{eq}}.
\end{aligned} \tag{V.17}$$

Here,

$$F_l \equiv 4 \int dx \{ f'_l(x) \}^2, \tag{V.18}$$

where $f'_l(x)$ denotes the Fourier transform of $i\xi f_l[\xi]$. For the case where $f_l(x)$ is twice differentiable, F_l can be rewritten as $F_l = -4 \int dx f''_l(x) f_l(x)$.

We finally prove Eq. (7) using Eq. (16). Putting $\hat{a}^0 = \hat{a}$ and $\hat{a}^1 = \hat{b}$, we have

$$\begin{aligned}
\overline{\Delta a_{\bullet}^0 \Delta a_{\bullet}^1} & = \int da_{\bullet}^0 da_{\bullet}^1 \Delta a_{\bullet}^0 \Delta a_{\bullet}^1 \langle f_0(\hat{a}^0 - a_{\bullet}^0) f_1(\hat{a}^1(t_1) - a_{\bullet}^1) f_1(\hat{a}^1(t_1) - a_{\bullet}^1) f_0(\hat{a}^0 - a_{\bullet}^0) \rangle_{\text{eq}} \\
& = \int da_{\bullet}^0 dx \Delta a_{\bullet}^0 \{ f_1(x) \}^2 \langle f_0(\hat{a}^0 - a_{\bullet}^0) (\Delta \hat{a}^1(t_1) - x) f_0(\hat{a}^0 - a_{\bullet}^0) \rangle_{\text{eq}} \\
& = \int da_{\bullet}^0 \Delta a_{\bullet}^0 \langle f_0(\hat{a}^0 - a_{\bullet}^0) \Delta \hat{a}^1(t_1) f_0(\hat{a}^0 - a_{\bullet}^0) \rangle_{\text{eq}} \\
& = \overline{\Delta a_{\bullet}^0 \langle \Delta \hat{a}^1(t_1) \rangle_{a_{\bullet}^0}}.
\end{aligned} \tag{V.19}$$

VI. RESULTS FOR THE CASE OF GAUSSIAN $f(x)$

A typical $f(x)$ is gaussian,

$$f(x) = \frac{1}{(2\pi w^2)^{1/4}} \exp \left[-\frac{x^2}{4w^2} \right] \tag{VI.1}$$

with $w = O(1) > 0$. In this section, we present results for this case.

The measurement error is simply given by

$$\delta a_{\text{err}} = w. \tag{VI.2}$$

Eq. (4) reads

$$\langle \Delta \hat{b}(t) \rangle_{a_\bullet} = \Theta(t) \frac{\langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}}{(w^2 + \delta a_{\text{eq}}^2)} \Delta a_\bullet. \quad (\text{VI.3})$$

Eq. (13) reads

$$\langle (\hat{b}(t) - \langle \hat{b}(t) \rangle_{a_\bullet})^2 \rangle_{a_\bullet} - \delta b_{\text{eq}}^2 = \frac{-1}{w^2 + \delta a_{\text{eq}}^2} \langle \frac{1}{2} \{ \Delta \hat{a}, \Delta \hat{b}(t) \} \rangle_{\text{eq}}^2 + \frac{1}{w^2} \langle \frac{1}{2i} [\hat{a}, \hat{b}(t)] \rangle_{\text{eq}}^2. \quad (\text{VI.4})$$

In Eq. (16), $\delta a_{\text{err}}^{j2}$ and F_j are given by

$$\delta a_{\text{err}}^{j2} = w_j^2, \quad (\text{VI.5})$$

$$F_j = 1/w_j^2 \quad (\text{VI.6})$$

for

$$f_j(x) = \frac{1}{(2\pi w_j^2)^{1/4}} \exp \left[-\frac{x^2}{4w_j^2} \right]. \quad (\text{VI.7})$$

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- [1] K. Koshino and A. Shimizu, Physics Reports **412**, 191 (2005), Chapter 4.
 - [2] R. Kubo, J. Phys. Soc. Jpn. **12**, 570 (1957).
 - [3] A. Shimizu and H. Kato, *Nonequilibrium Mesoscopic Conductors Driven by Reservoirs, Low-Dimensional Systems — interactions and Transport Properties* [ed. T. Brandes, Springer, 2000], pp.3-22.
 - [4] K. Fujikura, Master Thesis (University of Tokyo, 2015). [In Japanese].
 - [5] D. Goderis and P. Vets, Commun. Math. Phys. **122**, 249 (1989).
 - [6] T. Matsui, Rev. Math. Phys. **14**, 675 (2002).
 - [7] V. Jaksic, Y. Pautrat and C.-A. Pillet, Markov Process. Related Fields. **16**, 59 (2010).
 - [8] T. Terzioğlu, Mathematische Annalen. **182**, 236 (1969).
 - [9] L. Hörmander, *The Analysis of Linear Partial Differential Operators I: Distribution theory and Fourier Analysis* (2nd ed.). (Springer-Verlag, Berlin, 1990).
 - [10] M. Reed and B. Simon, *Methods of Modern Mathematical Physics: Functional Analysis I* (Revised and enlarged ed.). (Academic Press, San Diego, 1980).
 - [11] E. M. Stein and R. Shakarchi, *Fourier Analysis: An Introduction (Princeton Lectures in Analysis)*. (Princeton University Press, Princeton, 2003)
 - [12] E. H. Lieb and D. W. Robinson, Commun. Math. Phys. **28**, 251 (1972).
 - [13] B. Nachtergaele and R. Sims, Theorem. Commun. Math. Phys. **265**, 119 (2006).
 - [14] B. Nachtergaele, H. Raz, B. Schlein, R. Sims, Commun. Math. Phys. **286**, 1073 (2009).